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Best Available Techniques (BAT) for the Paint- and Adhesive Application in Germany

-Volume II: Adhesive Application-

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I Executive Summary

The Directive on *Integrated Pollution Prevention and Control (IPPC-Directive96/61/EG),* which was adopted by the Council of the European Community, has a significant impact on the permission of various industrial installations. As a main element for the transposition of the IPPC-Directive, according to article 16 passage two, an information exchange about the „Best Available Techniques“ is requested for all industrial sectors, listed in annex I of the IPPC-Directive, on an European level.

On this information exchange the member states of the European Community and also the affected industries participate. Content of the information exchange are: Specifications of the industrial sectors listed in the directive and the sector´s Best Available Techniques for an integrated prevention or reduction of environmental impacts, material consumption and emissions into the atmosphere, soil and water.

This final report represents the results which were examined from the Deutsch-Französischen Institut für Umweltforschung (DFIU-IFARE, German-French Institute for Environmental Research, University of Karlsruhe), on behalf of the Federal Environmental Agency (UBA). This report represents the German proposal for the Best Available Techniques for installations using organic solvents for surface treatment partition *paint and adhesive application* (cf. annex I, 6.7 of the IPPC-Directive) and will be submitted to the European BAT-documents.

I.1 Initial Position and Objective

Annex I. 6,7 of the IPPC-Directive defines the industrial activities via the consumption capacities of organic solvents. Industrial installations of the paint and adhesive application with a consumption capacity of 150 kg / h or 200 t / a, fall under the scope of the directive. This definition is different from those of other sectors, where specific plants are listed. Therefore relevant industrial activities had to be identified first.

Already in a very early project stage contacts, to manufacturers, authorities and industrial unions were established and organised in a working party.

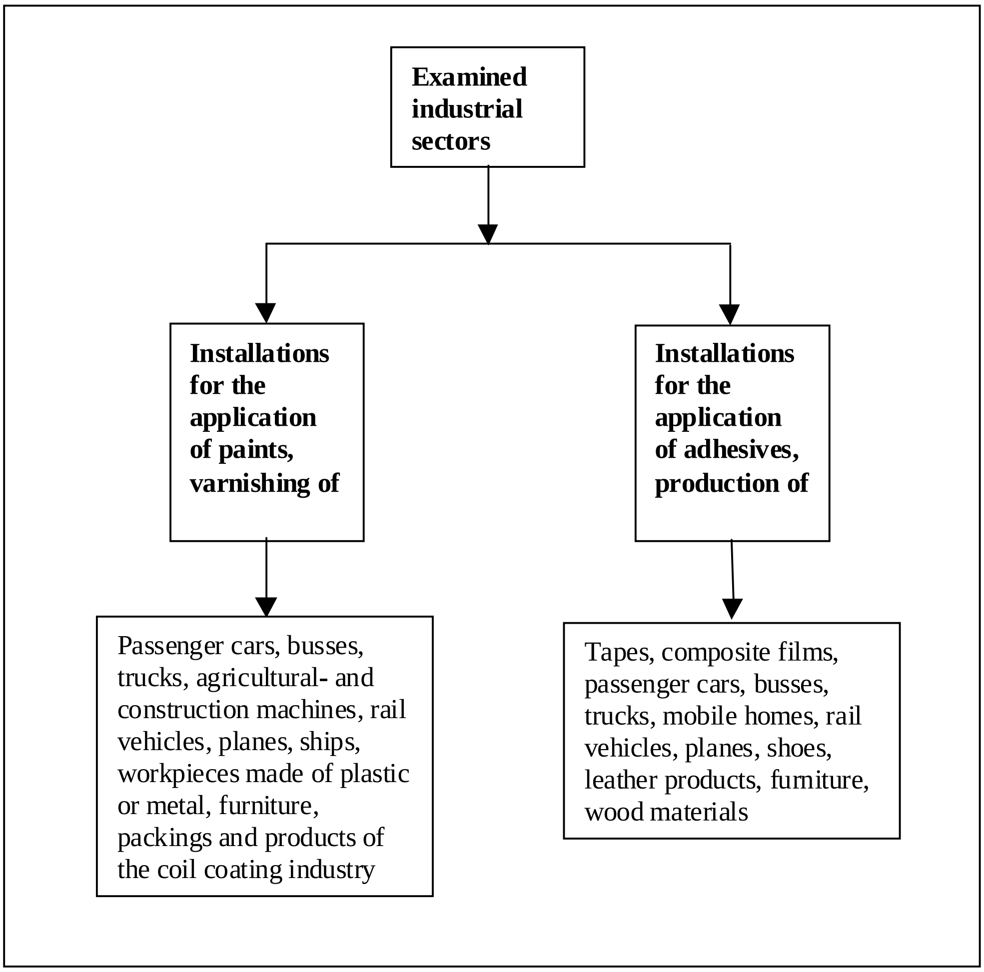

Figure I-1: *Investigated sectors of the industrial paint- and adhesive application*

Since there is only sparse current technical literature available for the problem, the process descriptions are based mainly on case studies of representative installations.
For the description of environmental relevant input- and output flows of enterprises, the applied processes were analysed via case studies. Thereby the process steps pre-treatment, coating and after-treatment were identified on basis of the production of typical workpieces in installations either relevant for the IPPC-Directive or that came under the scope before reduction measures were implemented.

Moreover technical options for a reduction or prevention of environmental pollution were characterised, if available. Main purpose of the case studies was the investigation of the following information:

- General information about the processes / plant
- Emissions into the atmosphere
- Water-side emissions
- Waste

- Material consumption and energy demand
- Other information such as data about waste heat or costs of applied measures.

Due to a keen competition in some sub-sectors, beside general process descriptions, no detailed data (cf. annex VI, volume paint application) could be obtained. Beside the case studies a further data collection from enterprises, was carried out via questionnaires (cf. annex VII). Processes such as drying, application or emission reduction measures, that are utilised within several sectors of the paint- and adhesive application, are sketched out in the annexes of the volume paint application.

On basis of the obtained data and informations, the *Best Available Techniques* were identified and as far as possible techno-economically analysed and integrated assessed.
The system boundary for the assessment is the specific plant. Environmental impacts, that result from previous processes within the supply chain, such as during the production of paints and adhesives, or during the products service life or its disposal were generally not regarded. But in the case of significant differences of environmental impacts between applied processes, these discrepancies were documented.

Due to the research project's support by the industry, legal authorities and industrial unions, it is ensured that a realistic description of the German installations is transmitted to the EIPPC in Sevilla.

The chapters were attuned with representatives from the industry, legal authorities and industrial unions in the context of workshops. During the project's runtime five workshops were organised by the DFIU. In the following, the case studies are listed.

Table I-1: *Case studies in the sector adhesive application*

Branch	Enterprise
Tapes	Astorplast Klebetechnik AG, Plant Alfdorf
	Tesa AG, Plant Hausbruch bei Hamburg
Paper and packaging, composite foils	CRYOVAC, Sealed Air Cooperation, Plant Flensburg
	Bischof + Klein GmbH & Co. KG, Plant Lengerich
Vehicles	Volkswagen AG, Plant Wolfsburg
	AUDI AG, Plant Ingolstadt
	MAN AG, Plant Salzgitter
	Ford Plant, Saarlouis
	MAN AG, Plant München
	EVO BUS, Plant Mannheim
	Daimlerchrysler, Plant Raststatt
	LHB-ALSTOM, Plant Salzgitter
	Hymer AG, Plant Grefrath
	Johnson Controls, Neustadt
	Johnson Controls, Grefrath
Shoes and leather goods	Salamander, Plant Kornwestheim
	RICOSTA, Plant Donaueschingen
	Gabor, Plant Rosenheim
	Schuh- und Lederwarenmesse, Pirmasens
	Navajo Dr. Genger Schuhfabrik GmbH, Hinterweidentahl
Wood materials and furniture	Schiedermöbel-Wohnmöbel GmbH & Co. KG, Plant Schieder
	ALNO AG, Plant Pfullendorf
	Dunlop AG, Plant Hanau

Table I-2: *Case studies in the sector of paint application*

Branch	Enterprise
Serial coating of passenger cars	Ford Plant, Köln
	Volkswagen Plant, Wolfsburg, Hannover und Emden
	DaimlerChrysler, Plant Raststatt
	Audi AG, Ingolstadt
	BMW AG, Plant Dingolfingen
	Verband der Automobilindustrie (VDA), Frankfurt
	Opel Plant, Eisenach
Coating of busses	EvoBus, DaimlerChrysler AG, Mannheim
Coating of trucks	MAN Plant, München,
	MAN Plant, München,
Coating of mobile homes	Hymer AG, Bad Waldsee
Coating of agricultural and construction machines	Claas Saulgau GmbH
Coating of rail vehicles	LHB-ALSTOM, Plant Salzgitter
Coating of planes	Lufthansa Technik
	Airbus, Hamburg
Coating of ships	Ecco-Paint Workshop, Hamburg
	Jos Lambert GmbH Meyerwerft, Papenburg
	Bremerhafener Dockgesellschaft, Bremerhafen
	Blohm+Voss Repair GmbH, Hamburg und JAFO Technologie, Hamburg
Coating of plastic workpieces	Dynamit Nobel AG, Plant Sterbfritz ECCA,
Coating of metal workpieces	Fachverband Bandbeschichtung VAW Aluminium AG, Grevenbroich
Coating of packings	Verband Metallverpackungen, Düsseldorf Rexam, Berlin
Coating of furniture	Schiedermöbel GmbH, Plant Schieder Alno AG, Pfullendorf

I.2 Results of the Determination of Best Available Techniques for the Industrial Paint Application

The consumption of environmental sound products such as powder coatings, water-dilutable paints and radiation curing materials is increasing over the last years, while the demand of solvent based paints is declining. This trend was also intensified by the transposition of the 31. BImschV[1] (Federal Emission Control Ordinance). In some sectors such as the coil coating industry or the serial varnishing of commercial vehicles, still mainly solvent based products are in use. In these sectors, the legal restrains are mainly achieved by thermal combustion units (cf. annex V, volume paint application). Biological processes for waste gas cleaning are only utilised against odours in the sectors of paint and adhesive application.
Due to the increasing utilisation of thin films and improved application techniques in all sectors, generally smaller material consumption can be observed.

Descriptions of generally utilised processes in the paint and adhesive application are documented in the annexes of the volume paint application:

Annex I: Possibilities of Energy Savings in the Paint Application

Annex II: Overview of selected Pre-treatment Processes

Annex III: Paint application

Annex VI:Drying

Annex V: Secondary Emission Abatement Measures for Reduction of VOC Emissions

I.3 Results of the Determination of Best Available Techniques of the Industrial Adhesive Application

Due to a strong substitution of solvent-based adhesives and to a lower extent due to migration of companies to low-wage countries, there are only a few industrial installations existing in Germany with a solvent consumption capacity of 150 kg/h or 200 t/a originating from adhesives applications and surface preparations. Installations falling under the scope of the Directive exist only in the sectors of production of adhesive tapes and in the paper and packaging industry for the production of compound foils. For these sectors, solvent-free products are not available for all applications at present. The emission limits are achieved via downstream located emission abatement measures (cf. annex V, volume paint application).
Due to the multitude of adhesives applied within the industrial applications, this sector is very complex.

[1] Ordinance for the transposition of the Directive 1999/13/EG

I.3.1 Production of Tapes

The selection of an adhesive system depends on the requested technical properties of the tape.

Table I-3: *Applications of pressure-sensitive adhesives for adhesive tapes* [KREUTER 2001]

Operative range or adhesive tapes	Application	Pressure-sensitive adhesive
Packaging	Adhesive labels, adhesive tapes for packaging	SIS block polymer as hot melt, ***solvent-based natural rubber adhesives (SB),*** water based acrylate dispersions
Paper industry	Splicing of paper rolls, transfer adhesive tapes	water soluble polyacrylates
Automotive industry	Transfer adhesive tapes for decorative strips and flank protection strips , insulating slabs, assembly aids for wheel case covers and side skirt pannels, adhesive tapes for winding of wire harnesses	***Solvent-based natural rubber adhesives, esters of acrylic acid and acrylates***
Construction industry	Foamed plastic adhesive tapes for sealing of windows, for caulking of construction splices and expansion joints, carpeting adhesive tapes etc	***Acrylate dispersions based on solvents*** or as watery dispersion
Medicine	Medical tapes, adhesive plasters	***Pure acrylates, polyacrylates***
Furniture industry	Transfer adhesive tapes for mirrors etc.	***Acrylate adhesives, solvent containing*** or watery

A determination of BAT for all products of the sector is therefore not possible. The required technical properties of the tape determine the BAT:

- Hot melts and dispersion for the lower quality range of packaging and masking tapes as well as double-sided tapes.
- UV cross-linking systems for solvent-based adhesives up to the lower quality range of transfer and packaging tapes as well as masking tapes.
- Solvent-based adhesive bulks for adhesive tape applications that require a high stress resistance. Up to now, there are no solvent-free adhesive bulks available that show equivalent properties. Therefore, there is no alternative to solvent-based adhesives in the high quality range. For these applications, techniques such as adsorption

installations and inert gas dryers followed by condensation installations are available that reduce emissions to the environment and allow a re-use of solvents. The emission limits, defined by the German 31 BImschV, are achieved.

Detailed data for the production of adhesive tapes are listed in Annex A.

I.3.2 Paper and Packaging Industry: Production of Compound foils

Solvent based adhesives are almost exclusively utilized for the production of composite foils. The production of composite foils for food packaging has the highest proportion in the lamination of composite foils. Within this sector the production of snack food packages is of utmost significance [VDI 1512]. Due to small amounts of adhesives used in the production of industrial packages, only production of consumer packages will be discussed in the following Due to the high variety of used materials and a similar construction of application devices, the selection of best available techniques is orientated best on the used adhesive system.

- For mass composites without stricter requirements the application of solvent-free adhesives is an already established BAT.
- Application of solvent-based adhesives with high solids contents (High Solid Systems) for the high performance sector for mechanically, thermally or chemically stressed packages and in the production of composites changing in material within one lamination installation. Subsequent exhaust gas cleaning techniques such as absorption and adsorption allow a high proportion of solvents to be re-used after distillation. For non-recyclable solvents (e.g. solvent mixtures), thermal or catalytic exhaust gas cleaning can be used (see also [VDI 2587/3]). Concentrations of less than 20 mg C/m³ in the clean gas are achieved.
- Reduction of fugitive emissions by distillation of solvent containing adhesive leftovers and cleaning solvents followed by recycling of the solvents.

II General Information

II.I Subject of the document

II.I.1 Context

On September 24th, 1996 the Council of the European Union adopted the Directive on Integrated Pollution Prevention and Control (IPPC Directive, 96/61/EG [IVU-RL 1996]). This Directive aims at harmonizing the permitting procedures for the operation of industrial installations [BECKER 1997]. Consequently, the Member States of the European Union will be committed to an "integrated concept" when licensing installations contributing to a certain extent to environmental pollution [RENTZ 1998]. According to Article 1, the purpose of the IPPC Directive is the achievement of a high level of protection of the environment taken as a whole. It lays down measures designed to prevent or, where that is not practicable, to reduce emissions in the air, water and land from certain industrial activities. Annex I of the IPPC Directive lists a series of industrial activities that are concerned by the Directive, especially ***"installations for the surface treatment of substances, objects or products using organic solvents, in particular for dressing, printing, coating, degreasing, waterproofing, sizing, painting, cleaning or impregnating*** **" (Annex I, No. 6.7).**

II.I.1.1 Environmental Hazard

Due to the generation of emissions of volatile organic compounds[1] (VOC), the mentioned installations show a substantial potential of environmental hazard. As a consequence of their neurotoxical properties, the emitted organic compounds lead also directly to a damage human health. After their release into the atmosphere, VOC are subject to photochemical reactions in the presence of nitrogen oxides (NOx) and sunlight, leading to the formation of photooxidant substances. Besides PAN (peroxyacetyl nitrate) and aldehydes, the leading photooxidant substance is the tropospheric ozon that mainly causes photochemical smog (so called "summer smog").

[1] Volatile organic compound (VOC) means any organic compound having a vapour pressure of 0,01 kPa or more at 293,15 K or having a corresponding volatility under the particular conditions of use [LM-RL 1999].

II.I.1.2 Significance of the IPPC Directive

The instrument for transposing the purposes of the IPPC Directive relies on the allocation of permits for the operation of industrial installations (new and existing installations). In the framework of permitting procedures, this Directive foresees substantial renewals with regard to former approaches dealing with environmental protection, amongst other things with regard to Directive 84/360/EWG that will be replaced by the IPPC Directive. The most significant modification and further development is the integrated approach of environmental protection through a cross-media contemplation taking into account the dangers of a displacement of environmental pollution from one media into another (cf. Article 9 (3) and 9 (4)) [BECKER 1997, RENTZ 1999].

The practical transposition of the IPPC Directive is realized through the application of "Best Available Techniques" (BAT). BAT shall serve for setting emission limit values for certain pollutants that are listed in Annex III of the Directive and for installations submitted to the IPPC Directive.

II.I.1.3 Definition of the Term "Best Available Techniques"

According to Article 2, section 11 of the IPPC Directive, 'best available techniques` shall mean *"the most effective and advanced stage in the development of activities and their methods of operation which indicate the practical suitability of particular techniques for providing in principle the basis for emission limit values designed to prevent and, where that is not practicable, generally to reduce emissions and the impact on the environment as a whole":*

- *'techniques` shall include both the technology used and the way in which the installation is designed, built, maintained, operated and decommissioned;*

- *'available` techniques shall mean those developed on a scale which allows implementation in the relevant industrial sector, under economically and technically viable conditions, taking into consideration the costs and advantages, whether or not the techniques are used or produced inside the Member State in question, as long as they are reasonably accessible to the operator;*

- *'best` shall mean most effective in achieving a high general level of protection of the environment as a whole.*

When determining best available techniques, all considerations that have already been described when assessing BAT-candidates, have to be taken into account and characterized, especially (according to Annex 6 of the IPPC Directive):

1. The use of low-waste technology;
2. the use of less hazardous substances;
3. the furthering of recovery and recycling of substances generated and used in the process and of waste, where appropriate;
4. comparable processes, facilities or methods of operation which have been tried with success on an industrial scale;
5. technological advances and changes in scientific knowledge and understanding;
6. the nature, effects and volume of the emissions concerned;
7. the commissioning dates for new or existing installations;
8. the length of time needed to introduce the best available technique;
9. the consumption and nature of raw materials (including water) used in the process and their energy efficiency;
10. the need to prevent or reduce to a minimum the overall impact of the emissions on the environment and the risks to it;
11. the need to prevent accidents and to minimize the consequences for the environment;
12. the information published by the Commission pursuant to Article 16 (2) or by international organizations.

II.II Relevant German and International Environmental Legislation

The national and international legislation concerning industrial solvent application (amongst other things) existing besides the IPPC Directive, will be described in the following. This part gives a short overview over relevant environmental legislation on international and German level concerning the given issues.

II.II.1 International Commitments

As international commitments for varnish-processing installations, the UN/ECE Protocols under the Convention on Long-range Transboundary Air Pollution, the European Directive on National Emission Ceilings (NEC Directive) and the European Directive on the Limitation of Emissions of Volatile Organic Compounds in Certain Activities and Installations [LM-RL 1999] can be specified.

II.II.1.1 The Convention on Long-range Transboundary Air Pollution of the UN/ECE and their Protocols

The Convention on Long-range Transboundary Air Pollution (CLRTAP) under the United Nations Economic Commission for Europe (UN/ECE) is a milestone in the international environmental policy. The Convention has allowed the development of international legislation and the creation of the substantial frame for controlling and reducing the hazard of environment and human health generated by the transboundary air pollution. The Convention was signed of 34 States and the European Community in 1979. It was the first international legally binding instrument dealing with air pollution issues in a transboundary context. This Convention has laid down the general principles of international cooperation for the reduction of air pollution and has created the institutional framework to bring together research and policy.

The Convention was fulfilled through five specific Protocols. The youngest Protocol, *Protocol to Abate Acidification, Eutrophication and Ground-level Ozone* (also called Göteborg Protocol) was signed at the end of the year 1999. This Protocol sets emission ceilings for the year 2010 for the four air pollutants sulphur (SO_2), nitrogen oxides (NO_x), VOC and ammonia (NH_3). The emission ceiling for Germany amounts to 995 tons VOC for 2010, which corresponds to a reduction of 70 % when comparing with 1990. This emission ceiling has to be taken into account when transposing further international commitments (for example Directive 99/13/EG) in order to meet their objectives too.

Furthermore, emission limit values for those four pollutants and best available techniques were set for specific sectors in the Technical Annexes of the Göteborg Protocols. These can or have to be considered respectively when elaborating a national reduction plan (cf. [UN/ECE 2001]).

II.II.1.2 European Directive on National Emission Ceilings (NEC-Directive)

The Göteborg protocol of the UNECE is implemented for the air pollutants SO_2, NH_3, NO_x and VOCs in the European Union by the Directive 2001/81/EG about national emission ceilings (also known as NEC Directive). It defines obliging national emission ceilings for the single member states beginning with the year 2010. There are a few variances compared with the Göteborg protocol. Until October 2002, all member states have to develop plans about how to achieve the necessary emission reductions. In this respect, national authorities specify themselves which measures to be the most suitable in order to fulfill the emission ceilings set. The year 2010 represents only a preliminary target, for which the unprotected ecosystems have to be reduced by min. 50 % when comparing to the situation in 1990. The emission load for Germany shall be reduced to 995 kt until 2010 in Germany (cf. also [EU 2001]).

II.II.1.3 The Solvent Directive of the European Union

The Solvent Directive [LM-RL 1999] deals with the limitation of volatile organic compounds, arising from 21 relevant activities of solvent application. The Directive is subdivided into a general part with the essential requirements and a sector specific part, in which

- the activities are defined (Annex I),
- threshold and emission limit values for installations of the 21 activities are listed (Annex IIA),
- principles and requirements for a reduction plan on installation level are named (Annex IIB) and,
- guidelines for the elaboration of a solvent management plan on installation level are given (Annex III).

The implementation of the Solvent Directive into national legislation was carried out in August 2001 (cf. [MAHRWALD 2000]).
In comparison with the German Technical Instruction on Air Quality Control, the Solvent Directive also covers the largest part of installations dealing with varnish application that were not subject to permissions until now. A substantial constituent of the Solvent Directive is the Solvent Management Plan.

The Solvent Management Plan

The Solvent Management Plan represents a certain form of material accounting in which the plants account regularly for their input and output flows aiming at reaching an efficient input of materials. In this frame, individual solvents have to be expelled separately according to the classification principles of the law of chemical substances as carcinogenic, mutagenic, toxic for reproduction or very unhealthy. Authorities check the presented solvent management plans with regard to completeness and validity, and, if necessary, agree upon precise reduction objectives with the plant [RENTZ 2001, MAY 2001]

II.II.2 Legal Bases in Germany

The essential national requirements for the adhesive and varnish processing enterprises will be listed shortly in the following. Important legal bases for industrial installations in Germany are defined in the following laws and ordinances:

- Federal law for the protection against emissions (BImSchG)
- Ordinance for the implementation of the Directive 1999/13/EG about the limitation of VOC emissions (31.BimSchV)(March 2001)
- Law on lifecycle management and waste (KrW-/AbfG)

- Water resources law (WHG)

For a permission of industrial installations, all damages to all environmental media are considered in Germany. While special emphasis lays on emissions, noise exposure and others are considered as well. As the precautionary principle is applied, limit values for the pollution are defined and their fulfillment is monitored through the authorities dealing with the permissions. The implementation of environmental laws and ordinances falls under the scope of the provinces. Hence emission limits may be different depending on the province. Furthermore, execution of an environmental impact assessment (UVP[2] law) is required for the permission for the construction of new industrial installations having a potential of environmental hazards.

II.II.2.1 Laws

II.II.2.1.1 Federal Law for the Protection against Emissions (Bundesimmissionsschutzgesetz, BImSchG)

The Federal law for the protection against emissions (Bundesimmissionsschutzgesetz, BImSchG) represents the basic principle and the TA Luft (Technical Instructions on Air Quality Control) the technical administrative instruction with regard to clean air act in Germany. This law formulates targets for installations and deals with principles and sequences of permitting procedures as well as with punishable acts and irregularities. The TA Luft is mainly used by permitting authorities as an orientation valid for the whole republic, whereby the limit values defined via the objectives of § 1 of BImSchG shall be supervised. Besides the documentation of the state-of-the-art, the TA Luft also formulates new principles such as redevelopment of existing installations, dynamisation, mass limitations and others (cf. [MAY 2001]).

II.II.2.1.2 Law on Lifecycle Management and Waste (Kreislaufwirtschafts- und Abfallgesetz, KrW-/AbfG)

According to the Law on lifecycle management and waste (Kreislaufwirtschafts- und Abfallgesetz, KrW-/AbfG), all plants exceeding an annual generation of 2 tons of waste needing special monitoring or 2 000 tons of waste needing monitoring, respectively, a waste management concept (§19) and a waste balance (§20) have to be compiled. For that purpose, information on type, amount and disposal of these wastes have to be collected. The compilation of a *waste management concept* on plant level was required for the year 1999 for a period of 5 years for the first time and shall include details on [MAY 1997]:

- type and amount of generated waste,
- implemented and planned measures for waste reduction, recycling and disposal,

[2] UVP is the German abbreviation of Environmental Impact Assessment

- reasons for the necessity of waste disposal,
- ways to recycle and dispose off waste,
- planning of sites and installations when waste disposal on-site, and
- remaining verification when disposing waste inside or outside Germany.

On the other side, the *waste balance* represents a retrospective description of the passed year with regard to waste management, whereby items 1, 3 and 6 of the above mentioned enumeration are to be fulfilled. Corresponding balances had to be elaborated first in 1998 retrospectively for the year 1997.

II.II.2.1.3 Water Resources Law (Wasserhaushaltsgesetz, WHG)

The water resources law (Wasserhaushaltsgesetz, WHG) builds the basis for the protection of ground water and surface water. According to this law, wastewater containing dangerous substances has to be treated in compliance with the state-of-the-art before being drained. For that purpose, the dangerous substances are classified with regard to their degree of water hazardousness (water hazard classes): solvent-based paints correspond to class 2 (WGK 2) (hazardous) and water-based paints to class 1 (WGK 1) (slightly hazardous). The corresponding information is documented on security data sheets for paints and solvents.

The water resources law is completed by administrative instructions for 57 branches, in which emission limit values for certain substances and parameters have been set. Unification is being performed step-wise, flowing into the TA Wasser that corresponds to a general framework of administrative instructions dealing with the minimal requirements for the dumping of wastewater into surface waters. In addition to the requirements set via limit values, the state-of-the-art is formulated through general requirements (cf. [MAY 2001]).

II.II.2.2 Ordinances

II.II.2.2.1 Federal Ordinances for the Protection against Emissions (Bundesimmissionsschutzverordnungen, BImSchV)

Since the targets of the Federal law for the protection against emissions are not sufficient for precise questions dealing with technical planning, 31 Federal Ordinances for the Protection against Emissions (Bundesimmissionsschutzverordnungen, BImSchV) have been adopted. Concerning installations for surface treatment using organic solvents, the following ordinances are of special importance (cf. [MAY 2001]):

- 2nd Ordinance (2. BImSchV): Halogenated hydrocarbons (March 1991)
- 4th Ordinance (4. BImSchV): Installations submitted to permitting (November 1985)
- 12th Ordinance (12. BImSchV): Ordinance on plant failures (September 1991).

- 31st Ordincance (31. BImSchV): Implementation of the EU Directive 1999/13/EG of March 11th, 1999 (cf. II.2.2.3)

II.II.2.2.2 Ordinance Concerning Dangerous Substances (Gefahrstoffverordnung, GefStoffV)

According to § 16, section 3a of the Ordinance concerning dangerous substances (Gefahrstoffverordnung, GefStoffV), the operator is committed to carry out an inventory of all dangerous substances used within the installation. This inventory shall include information on notations, designations and amounts of the dangerous substances within the installation as well as on the departments where dangerous substances are used [MAY 1997]. This ordinance is not oriented towards thresholds, but the elaboration of balances is principally required.

II.II.2.2.3 Ordinance for the transposition of Directive 1999/13/EG related to the limitation of VOC-emissions (31.BimSchV)

This ordinance is the implementation of the EU Solvent Directive into national legislation [31. BIMSCHV 2001]. In this regulation, single requirements of the EU Solvent Directive will be implemented more strictly than it was necessary according to EU instructions. The superior target is achievement of the maximum amount of emissions of 995 tons for the year 2010 as specified in the Göteborg protocol.

II.III Subject of the Research Project

Within the scope of the implementation of the IPCC Directive, this report about the best available techniques for the sectors of industrial varnish and adhesive application has been created. This report is the German contribution to the European BAT Data Sheets for the named sectors. The Institute for Prospective Technological Studies (IPTS) in Sevilla manages the creation of the European BAT Data Sheets for all sectors. According to the schedule, works on the BAT Data Sheets “Surface Treatment with application of organic solvents for the sectors varnish and adhesive application” are supposed to begin in spring 2002[3]. The German Federal Environmental Agency (Umweltbundesamt) is in charge of the coordination of activities on national level on behalf of the German Government.

Due to extensive literature research of technical literature and numerous expert discussions with industry representatives and representatives of industrial associations, the sectors with maximum solvent consumptions have been selected. These have been investigated for the creation of this report (cf. Table II-1).

The product spectrum of the industry sectors shows a very high variety resulting in strongly different requirements for adhesive bondings and varnish coatings. Hence production processes and applied techniques of the investigated sectors also strongly differentiate, so that they can only be described separately. For the task of a definition of the BAT candidates, a

[3] At the beginning of the project, the start of TWG was supposed to be in summer 2000

detailed description of the applied techniques is necessary in order to prevent if misinterpretations in the further creation of BREFs (cf. [IPCC 2002]).

Table II-1: *Investigated industrial sectors of varnish or adhesive processing*

Adhesive processing installations for production of:	**Varnish processing installations for coating of:**
Adhesive tapes	Passenger cars
Composite foils	Buses and trucks
Passenger cars	Mobile homes
Commercial vehicles: buses, trucks	Large vehicles
Mobile homes	Rail vehicles
Rail vehicles	Aircrafts
Aircrafts	Ships
Shoes and leather goods	Plastic parts
Wood materials and furniture	Furniture
	Metal work pieces
	Products of the coil coating industry
	Packages

In order to describe the input and output flows that are environmentally relevant, the technical processes implemented nowadays are to be submitted to a comprehensive analysis. The investigated production systems are very heterogenous sectors that process a few thousand materials. As there is only a little technical literature dealing with such issues, the process descriptions are based on case studies and enterprise inspections of representative installations. The process steps pretreatment, coating and post-treatment will be identified by means of the cycles of typical work pieces in IPCC relevant installations or installations that have been ICPP relevant installations before the invention of abatement measures. Also the associated mass and partly energy flows are described. Furthermore, technical options for avoidance and reduction of environmental pollution are characterized if existing. The target of the case studies was the achievement of the following information:

- general information on the installation/the process
- emissions into air
- emissions into water
- waste

- input material and energy consumption
- further data such as details on waste heat, eventually details on cost aspects of implemented emission reduction measures.

Based on the acquired data and information, the Best Available Techniques (BAT) have been identified and as far as possible analyzed with respect to technical economic aspects and assessed under consideration of cross-media aspects.
In doing so, the area of balancing for the cross-media assessment is the respective installation. Environmental impacts generated in pre-chains of the considered production processes cannot be considered generally due to the complexity of products and processes. For example such impacts are generated by the production of varnishes and adhesives, during the usage period and disposal of the product. For sectors where there are significant differences with respect to the environmental impact outside of the investigated installations this has been pointed out.

With the support of the research project by industry, associations and permitting authorities, it can be ensured that the efforts are optimized and a realistic picture of the German installations is conveyed to the EIPPCB in Sevilla.

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III Adhesive Processing

For industrial applications, the joining technique gluing has increased significant over the last years. A joining of different lightweight construction materials (such as in vehicle construction) can often not be achieved by classic joining techniques. Combination of certain materials is only enabled by adhesive techniques. In contrary to welding connections, there are no structural changes resulting in a reduction of material strength at the joining point [PICKER 2000]. Further, the adhesive junction allows a homogenous force flow and hence a good strength behaviour even for low material thickness [INDUSTRIEVERBAND KLEBSTOFFE 2000]. The junction of components by adhesives shows the following advantages if compared to other joining techniques:

- Different work pieces can be joined that cannot be combined by classic joining techniques (such as welding).
- The specific material properties of the work pieces to be joined are maintained.
- Adhesives allow for novel constructions with integration of important functions (e.g. electrically conducting adhesives).
- Optically immaculate surfaces of the joined components can be achieved by application of adhesives.

Besides the use as joining materials for components, adhesives are also used as sealants in technical applications. Sealants create a connection between the surfaces of the work pieces [STACHOWAIK 1998].

Adhesives

According to DIN 16920, an ***adhesive*** *is a non-metal material being able to join adherends by surface adhesion and inner strength. The junction is a result of cohesion and adhesion forces.*

Sealants

According to DIN 52460, a ***sealant*** *is a non-metal material filling gaps (splices. cavities) gas and fluid tight between compounds due to material conclusion and volume bridging properties. The structure of the compounds does not change essentially.*

Often a clear assignment of a technological application to an application purpose is not possible, as the used adhesives systems both connect (join) the compounds and seal them against influences from outside. The application "joining" is predominant for the sectors. Adhesives and sealants are therefore cited as adhesives and will be discussed together in the following.

III.1 Adhesive Consumption

In 1999, 509,000 tons of adhesives and 123,000 tons of sealants were produced in Germany. For comparison, 340,000 tons of adhesives were produced in 1995 [HEßLAND 1997, INDUSTRIEVERBAND KLEBSTOFFE 2000].

The produced adhesives can be sorted by the adhesive systems illustrated in figure III-1.

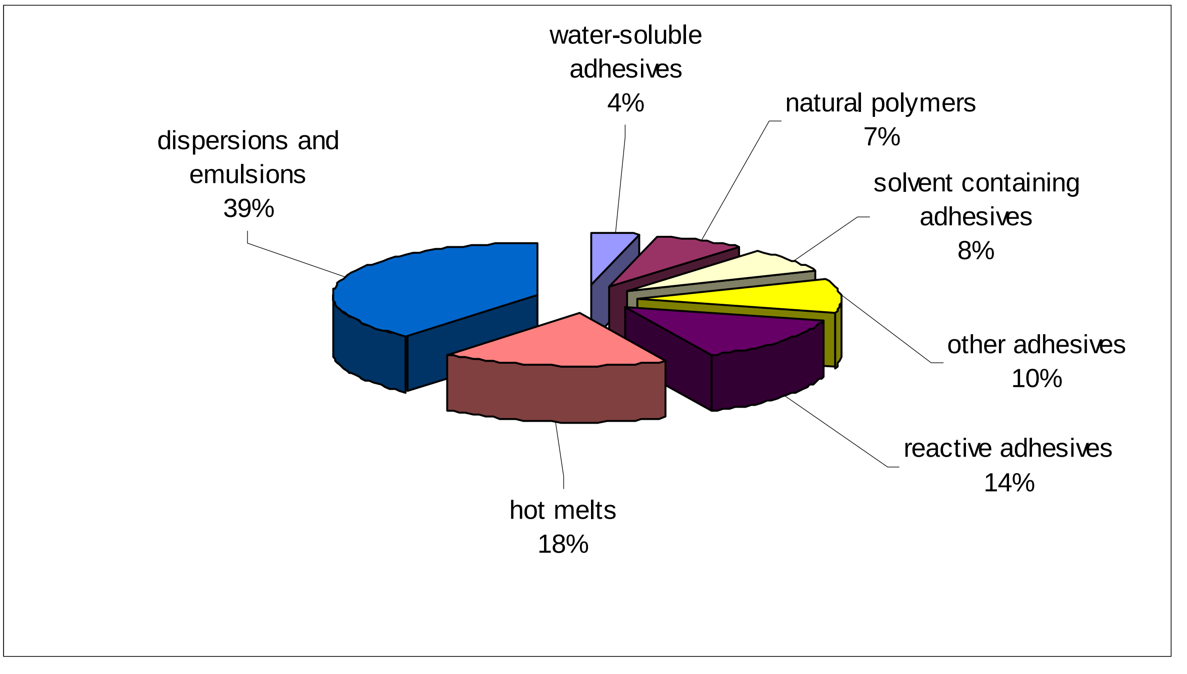

Figure III-1: *Proportions of the total amount of the adhesives produced in 1999*

The dispersion and emulsion adhesives have the highest proportion (30 %) of the adhesives produced in 1999. The carrier medium for these adhesive systems is water. Solvent-based adhesive systems have a proportion of 8% of the total amount. Their proportion is further decreasing. Natural polymers are adhesives based of glutamine and caseine. Other adhesives shown in figure I-1 are solvent-free systems that cannot be assigned to the other groups (e.g. bitumen).

The solvents of industrial adhesive applications cause 7% of the total VOC emissions. The highest VOC emissions originate from varnish processing and other applications (plastic foaming, aerosols, cosmetics, plastic processing as well as machining of metals) (cf. volume "application of varnishes").

Organic solvents in the adhesive systems serve for the dissolution of the adhesive polymers and transform these into a low viscous state. This is for example necessary in order to achieve a sufficient wetting of the surfaces being joined. After the application of adhesives onto the surfaces being joined, the solvents evaporate and contaminate the environment as so-called volatile organic compounds (VOCs). Solvents of adhesive systems are alcohols, ketones, esters and aromatic hydrocarbons [STACHOWAIK 1998]. Typically the solvents exist as mixtures in the adhesive systems.

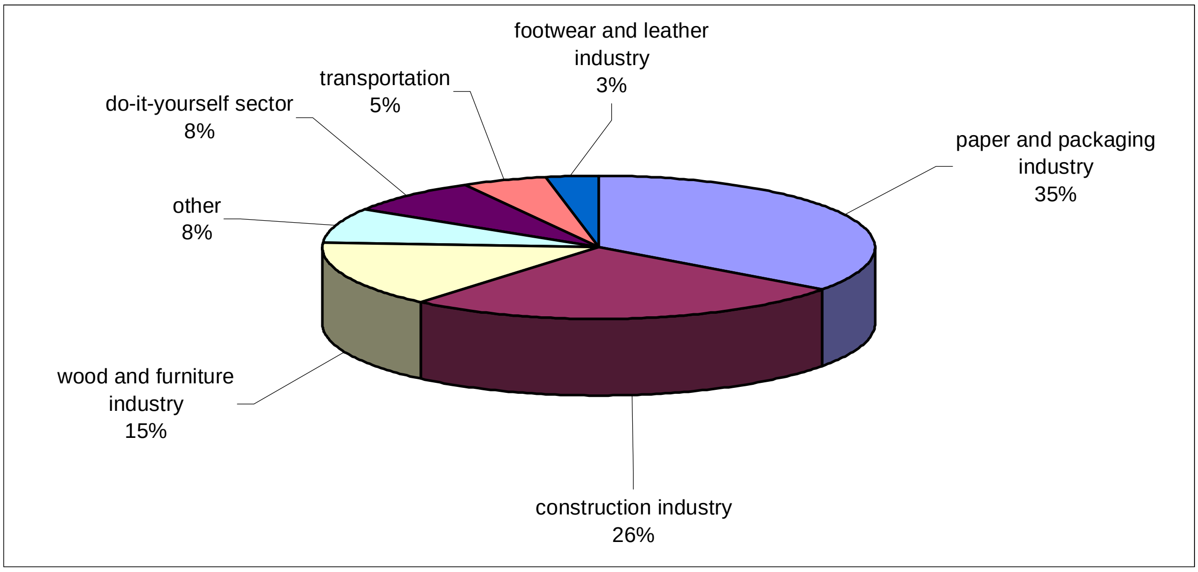

Figure III-2: *Adhesive consumption of the market segments* [INDUSTRIEVERBAND KLEBSTOFFE 2000]

The adhesive consumption in Germany can be assigned to different market segments as sketched out in fig III-2. The highest proportion of adhesive consumption has the paper and packaging industry (35%). Adhesive applications of the construction industry and the do-it-yourself sector is no industrial installations according to the Directive. Adhesive application in the construction industry is carried out predominantly by handicraft enterprises at the construction site. The do-it-yourself sector is no industrial installation and is therefore not investigated within this project. Assembly processes in electric engineering and machine industry as well as the production of adhesive tapes are summarized as other applications in figure III-2.

The application of solvent containing adhesive systems has been substituted by solvent-free adhesive systems in the most sectors due to environmental protection and production technology reasons. In 1995, the proportion of solvent containing adhesive systems was 12.5 % of the total amount produced in Germany and in 1999, it amounted only 8% (see fig III-1). For some industrial sectors, no substitution has taken place yet. Presently environmentally friendly products with equivalent technological properties are still missing. Fulfillment of law regulations is achieved by the use of subsequent exhaust gas cleaning installations. Especially post-combustion, adsorption and absorption techniques are used as secondary emission abatement measures.

III.2 Classification of the Adhesives

About 250,000 different adhesives are produced worldwide. In Germany, about 25,000 different adhesive products are available for trade. Due to the high variety of the compounds, a uniform and detailed classification of adhesives is not possible [HABENICHT 1997]. Hence, adhesives are normally classified by the chemical basis or the setting mechanism [STACHOWAIK 1998].

• Classification by the Chemical Basis

Organic and inorganic adhesives can be differentiated in the classification by the chemical basis (see figure III-3). The majority of the adhesives used in the technical sector are artificially produced organic compounds.

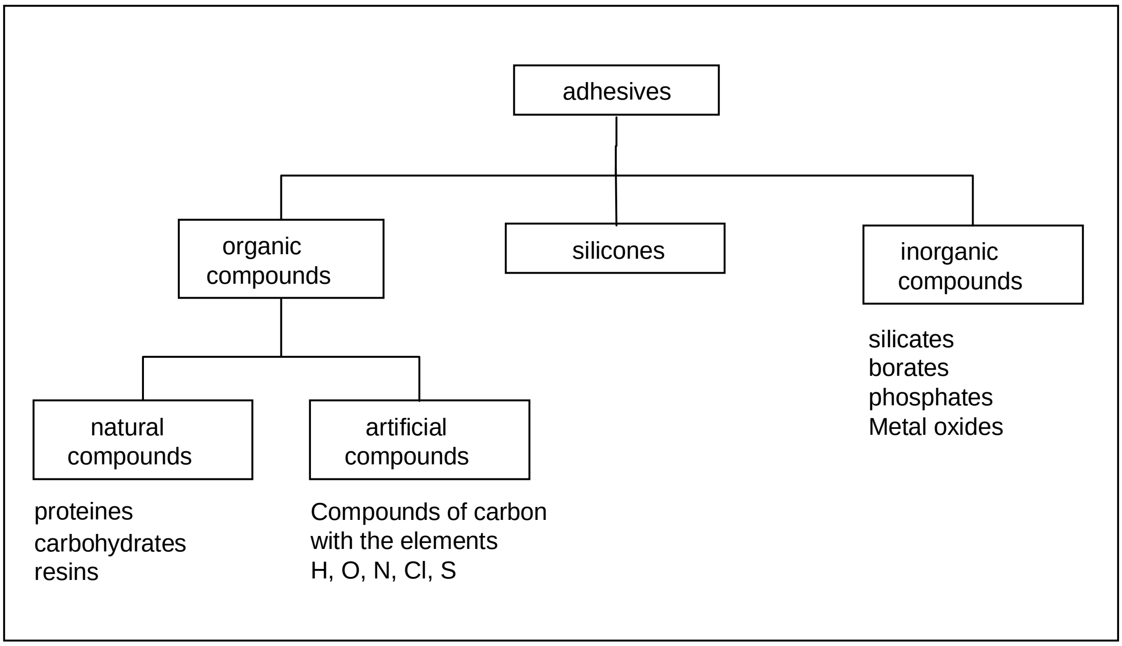

Figure III-3:*Classification of the adhesives by chemical basis* [ENDLICH 1995]

• ***Classification by Setting Mechanism***

If the classification is oriented on the setting mechanism, the chemically reacting systems and the physically reacting systems can be differentiated. An additional parameter of the classification is the number of components of the adhesive system. Classification of adhesives by setting mechanism is illustrated in figure III-4.

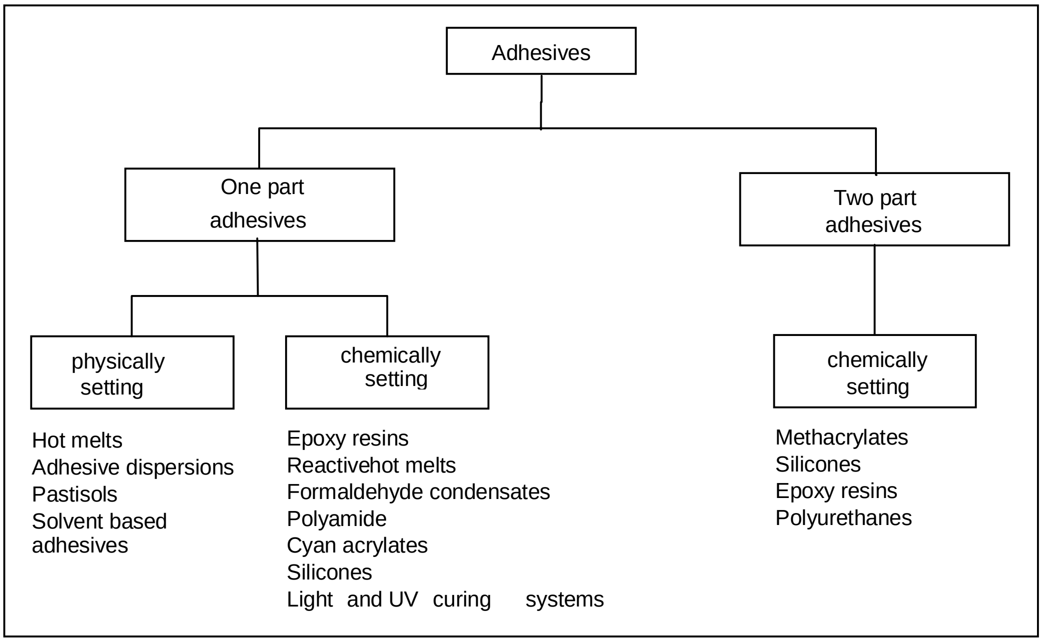

Figure III-4: *Classification of the adhesives by the setting mechanism*

• ***Chemically Reacting Systems***

Before the adhesion process, there are monomer and pre-polymer molecules ready for reaction in the adhesive layer. They react with each other under certain conditions (e.g. pressure or temperature) forming polymers in the adhesive splice. For chemically reacting systems, there are typically two reaction partners existing in a mixture that cause the reaction. The components are adhesive and hardening agent. Hence these adhesives are so-called **2 part adhesives**. The connection of the low-molecular compounds existing in the liquid adhesive with the high-molecular compounds existing in the adhesive layer after curing, results from different reaction mechanisms. Additives and filling materials are added to the chemically reacting systems in order to achieve certain technological properties.

For one part adhesives, the second component necessary for curing is already present in the adhesive splice (e.g. air humidity). The hardening of the reaction curing **two part** adhesives occurs due to chemical reactions between the hardening component and the adhesive. The chemical reactions leading to the curing are polymerization and polyaddition reactions. For example methyl methaacrylates are polymerization adhesives used in serial production. Polyaddition adhesives of this group are representatives of 2 part systems with epoxy and polyurethane compounds [STACHOWAIK 1998, HABE NICHT 1997, PICKER 2000].

The curing of **hot-setting materials** occurs due to heat-regulated polymerization. The temperatures applied vary depending on the compound between 100 and 250° C. For example polyaddition adhesives based on epoxy resins used in automotive shell construction are hot – setting materials.

For **moisture-setting materials**, the compounds exist as monomeres. The single molecules are able to form polymers due to several functional groups. These compounds are processed as one or two part systems. The curing reaction occurs after application due to diffusion or air humidity into the adhesives. Moisture-setting compounds are polymerization adhesives for example based on cyanacrylates as well as compounds belonging to the group of polyurethane adhesives and silicones [PICKER 2000]. For example they are used for junctions of glass with metal.

For **anaerobic curing materials**, the curing occurs due to polymerization after the joining of metals under the created oxygen-free conditions. Curing of the compounds can be accelerated by heat. Some of the methaacrylates used in automotive construction are anaerobic setting compounds [PICKER 2000].

• Physically Setting Systems

The physically setting systems consist of only one component – the polymers already existing in the liquid adhesive. These are supplied with additives and filling materials in order to improve their properties. The polymers are liquified by elevated temperatures, dissolved in a solvent or dispersed in water or solvent in order to achieve wetting capable conditions. Hot melts, adhesive dispersions and solvent adhesives belong to the group of physically setting systems.

Hot melts represent the biggest group of physically setting systems. They are thermoplastic compounds being solid at room temperature that cure by cooling of the melted material, where cohesion and adhesion forces are created simultaneously.

• Physically-Chemically Setting Systems

The reactive hot melts are a combination of physical and chemical mechanisms. The adhesives are applied in heated condition and for a sufficient handling strength after cooling followed by curing due to a chemical reaction [STACHOWAIK 1998].

III.3 Activities Falling under the Scope of the Directive

The following sectors process significant amounts of adhesives:

- Production of adhesive tapes
- Paper and packaging industry
- Automotive industry including the activities production of passenger cars, commercial vehicles, trailers and mobile homes, supplier parts, rail vehicles and aircrafts.
- Footwear industry
- Leather goods industry
- Wood and furniture industry
- Industrial assembly processes

Due to a strong substitution of solvent-based adhesives and to a lower extent due to migration of companies to low-wage countries, only a few industrial installations exist in Germany with a solvent consumption capacity of more than 150 kg/h or 200 t/a originating from adhesives and surface preparations. Installations falling under the scope of the Directive concerning the application of adhesives exist in the sectors production of adhesive tapes and paper and packaging industry in the production of compound foils. The application of solvents by installations of the investigated sectors is illustrated in table III-1.

Table III-1: Solvent use and relevance for the Directive of industrial installations of investigated sectors

Installations for production of:	Installations relevant for the Directive	Solvent consumption of 50 to 280 kg /day	Solvent consumption of 0 to 50 kg/day
Leather goods			+
Wood materials			+
Furniture			+
Products of machine industry and electrical engineering			+
Passenger cars			+
Commercial vehicles			+
Aircrafts		+	
Buses		+	
Rail vehicles		+	
Footwear		+	
Supplier parts for passenger cars			+
Adhesive tapes	+		
Packaging (compound foil)	+		

In the following, a detailed consideration of production processes is given for sectors including installations exceeding a consumption capacity of 150 kg/h or 200 t/a and best available techniques will be identified. For sectors that include installations with lower consumption capacities, the state-of-the-art will be described and alternative techniques for emission abatement of VOCs will be cited, if installations with higher consumption capacities are expected to exit in Europe. Sectors with installations processing only minor amounts of solvent adhesives and that do not have any significance for foreign countries are not studied further.

III.4 Case Studies in Adhesive Application

Due to the poor documentation of adhesive application in the specialist literature, this investigation is based on various case studies of industrial enterprises listed in the following table III-2.

Table III-2: *Case studies in adhesive application*

Sector	Enterprise	Contact
Production of adhesive tapes	Astorplast Klebetechnik AG, Werk Alfdorf	Mr. Gemünd, Herr Kreuter
	Tesa AG, Werk Hausbruch bei Hamburg	Mr. Weißleder, Herr Dr. Röber
Paper and packaging	CRYOVAC, Sealed Air Cooperation, Werk Flensburg	Mr. Kahl, Herr Becker
	Bischof + Klein GmbH & Co. KG, Werk Lengerich	Mrs. Lütke-Lengerich, Herr Reinisch
Transportation	Volkswagen AG, Werk Wolfsburg	Mr. Dr. Mrowitz, Herr Dr. Gruber, Herr Stege
	AUDI AG, Werk Ingolstadt	Mr. Dr. Achatz, Frau Kaulberg
	MAN AG, Werk Salzgitter	Mr. Jurek
	Ford Werke, Saarlouis	Mr. Dr. Buchem, Herr Dr. Clysters, Herr Meffert, Herr Gebert
	MAN AG, Werk München	Mr. Golling, Herr Dr. Schmid
	EVO BUS, Werk Mannheim	Mr. Janker, Herr Siener
	Daimlerchrysler, Werk Raststatt	Mr. Höhner

Sector	Enterprise	Contact
Transportation	LHB-ALSTOM, Werk Salzgitter	Mr. Dr. Neutzler, Frau Ziems
	Hymer AG, Werk Grefrath	Mr. Priebe
	Johnson Controls, Neustadt	Mr. Koch
	Johnson Controls, Grefrath	Mr. Dr. Prömper
Footwear and leather goods industry	Salamander, Werk Kornwestheim	Mr. Raith
	RICOSTA, Werk Donaueschingen	Mr. Allaut, Herr Kinzelmann
	Gabor, Werk Rosemheim	Mr. Herberger
	Navajo Dr. Genger Schuhfabrik GmbH, Hinterweidentahl	Mr. Farbacher
Wood and furniture industry	Schiedermöbel-Wohnmöbel GmbH & Co. KG, Werk Schieder	Mr. Krain, Herr Schouwenaars
	ALNO AG, Werk Pfullendorf	Mr. Fink
	Dunlopp AG, Werk Hanau	Mr. Dr. Wolfahrt

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1 Production of Adhesive Tapes

The threshold values for the consumption capacity of organic solvents regulated by the Directive (annex I; no 6.7) are strongly exceeded in the production of adhesive tapes, depending on the applied production method. There has already been a far-reaching substitution of solvent-based adhesives within some parts of the sector. However for products that require permanent high adhesive strength and cohesiveness, there is still no alternative to solvent-based products. For an emission reduction and recovery, secondary measures are used, such as adsorption techniques. The production of adhesive tapes was investigated in two enterprises.

1.1 General Information

In 1999, 690 million m² adhesive tapes were produced in Germany for packaging, vehicle production, electrical equipment, construction, furniture- and paper industries as well as for the so-called “do-it-yourself” sector [SBW 2000]. Adhesive tapes are widely used in the vehicle industry for example for attachment of parts onto outer surfaces of car bodies, for abrasion protection, for caulking openings, for wrapping cable harness, for masking during varnishing and for supporting functions during assembly. Depending on the use, different requirements concerning product properties such as adhesive strenght, resistance against temperature, mechanical rigidity and tensile strength have to be met [IKV 2000]. VOC emissions in the production of adhesive tapes originate from the application of solvent-based adhesive systems and the application of solvent containing auxiliary material such as release agents and primers [HENDEß 1998]. In 1995, 15.560 tons organic solvents were used as components in adhesive systems for the production of adhesive tapes in Germany. In 1997, emissions originating from processing of adhesives for production of adhesive tapes amounted 415 t/a. This is a small amount if compared to the processed quantities due to efficient exhaust gas treatment [BMU 1997].

1.2 Applied Processes and Techniques

1.2.1 Construction of Adhesive Tapes

Adhesive tapes consist of a substrate, a coupling agent, a pressure-sensitive-adhesive and releasing agents. As for substrates, especially paper, fabrics, polyvinyl chloride foils, polyester, polyamide and aluminium are processed. Due to the wide variety of products, a simplified classification seems reasonable for determination of the best available techniques. Products can be classified as transfer adhesive tapes (double sided adhesive tapes), adhesive tapes for packaging purposes and adhesive tapes for coating purposes.

- For production of **one-sided adhesive tapes**, a pressure-sensitive-adhesive is applied onto a substrate. Adhesion of the adhesive layer onto the substrate is improved by a physical surface preparation or an application of primers.
- For production of **double-sided adhesive tapes**, a pressure-sensitive-adhesive is applied onto both sides of the substrate (cf. Fig 1-1). A releasing agent is appliqued onto one of the adhesive layers in order to avoid contact between the adhesive layers.

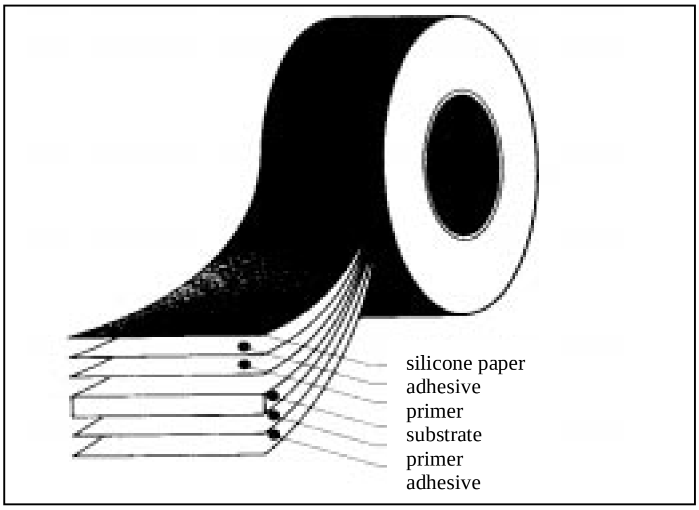

Figure 1-1*: Structure of double-sided adhesive tapes*

1.2.2 Materials for the Production of Adhesive Tapes

Pressure-sensitive-adhesives are organic compounds with a high self strength that have a high adhesive strength with different surfaces at the same time. Unlike other adhesive systems, the plastic adhesive film is also active in dry conditions. The adhesive shows a visco-elastic behavior and can be considered as fluid with very high viscosity [KREUTER 2001]. A classification of pressure-sensitive-adhesives is shown in table 1-1.

Table 1-1: *Pressure-sensitive adhesives for production of adhesive tapes*

Type of adhesive	**Caoutchouc adhesive**	**Acrylate adhesive**	**Other pressure-sensitive adhesives**
Dispersion in water	Styrene butadiene styrene block copolymer (SBS), SBR latex, carboxylated (CSBR)	Pure acrylate, polyacrylate	Polyvinyl acetate, acrylate, ethylene vinyl acetate (EVA)
Hot melt	Styrene isoprene styrene block copolymer (SIS), butadiene styrene copolymer (SBR), styrene butadiene styrene block copolymer (SBS)	Polyacrylate	No application
Dissolution in organic solvents	***Natural rubber (NR), butadiene styrene (SBR), styrene isoprene styrene block copolymer (SIS)***	***Pure acrylate, polyacrylate***	***Polyurethane, neoprene***
Systems with 100 % solids content, radiation curing or as two part system	Natural rubber (NR), butadiene styrene copolymer (SBR), styrene isoprene styrene block copolymer (SIS)	Pure acrylate, polyacrylate	Polyurethane, polyester, two part polyurethane

Solvent containing products are printed cursive

Caoutchouc Adhesives and Resins

Caoutchouc adhesives that are technically processed are natural rubber (NR) and synthetic caoutchouc adhesives such as butadiene styrene (SBR), styrene isoprene styrene (SIS) or SBR latex (carboxylated).

For production of **natural caoutchouk systems**, caoutchouc is dissolved in organic solvents together with resins influencing cohesiveness and adhesion (so-called tackifiers).
Other aggregates, such as agents preventing aging processes are also added.
These adhesives are mainly used for band-aids, but also for double-sided adhesive tapes, adhesive tapes for packaging and coating adhesive tapes [RÖBER, WIEßLEDER 2001].

Synthetic caoutchoucs are appliqued as hot melts without solvents. For production of such synthetic caoutchoucs, resin and other auxiliary materials are mixed in the melt, for example in continuously working extruders and then coated as a melt. The thermoplastic behavior of the compound is reversible [KREUTER 2001].

Acrylate Adhesives

Adhesive tapes are coated with acrylate adhesives especially for long-term or outside use. Further, acrylate adhesives are predominant in the field of assembly application due to their higher thermal and chemical resistance as well as higher cohesion if compared with the caoutchouc adhesives. The adhesives are appliqued as water based dispersions or/and dissolved in organic solvents (e.g. benzene, ethyl acetate or toluol). They can also be appliqued as 100% solid system (as UV cross-linking compounds) [KREUTER 2001]. Within this group, the solvent-based adhesives possess the highest cohesiveness [RÖBER, WEIßLEDER 2001].

Other Pressure-sensitive adhesives

Thermoplastic elastomers, polyurethanes, polyacrylates as well as silicones are also used for the production of adhesive tapes for special applications [RÖBER 2001].

Auxiliary Materials

Coupling agents allow a failure-free unwinding and re-stripping of the adhesive tape by increasing the adhesion between pressure-sensitive-adhesive and substrate. Common low emission techniques for surface treatment are corona discharge, flamming and low pressure plasma surface preparation. The coupling can also be realized by application of a primer layer. Primers are appliqued dissolved in organic solvents or as water based dispersions.

Releasing agents

Releasing agents are supposed to prevent adhesion of the pressure-sensitive adhesive to the surface of the adhesive tape. Silicones are appliqued as releasing agents for acrylate adhesives and synthetic caoutchoucs.

For natural caoutchuc adhesives, systems containing stearyl groups are in use additionally. Most of the releasing agents are applied as solution [HENDEß 1998]. Exceptionally water based and 100 % systems are processed [RÖBER, WEIßLEDER 2001].

Impregnating agents

Impregnating agents are appliqued for surface refinement of smooth, absorbent papers in order to increase the wet strength and mechanical rigidity. For this process, water-containing polymer dispersions are used based on SBR, acrylate and also acrylate–natural caoutchouc systems for special applications.

Solvents

Solvents for pressure-sensitive-adhesives are aromats (especially toluol), aliphatics and benzine as well as mixtures of these components. Solvent proportions in adhesives ready for application range between 75 and 60 %. Partly, high solid systems with solid proportions over about 60% are in use [KREUTER 2001].

1.2.2.1 Properties of Pressure-sensitive Adhesives

Table 1-2: *Properties of pressure-sensitive adhesives* [KREUTER 2001]

Parameter	Dispersions	Hot melts	UV cross-linking acrylates and electron beam curing systems	Solvent-based adhesives
Solvents	Water as carrier, problematic drying	No solvents	No solvents	Organic solvents, troublesome storage and drying, exhaust gas treatment, explosion protection
Sheeting velocity	Up to 250 m / min	Fast cross-linking, sheeting velocity up to 400 m / min	Very fast cross-linking, high sheeting velocity up to 450 m / min for electron beam curing	Up to 250 m / min
Cohesion/adhesion	Good cohesion and adhesion	High cohesion and low adhesion	High cohesion and adhesion (< than of solvent-based adhesives)	Highest Cohesion (due to maximum length of polymer chains) and adhesion
Cross-linking	Physical cross-linking	Physical cross-linking	Chemical cross-linking	Chemical cross-linking
Solids content	50 – 70 %	100 %	100 %	20 – 35 %
Aging resistance	Resistant against aging	low aging resistance and low resistance against UV	Resistant against aging	Resistant against aging
Temperature resistance	Temperature resistant	Low temperature resistance (50 – 100 °C)	Temperature resistant	Temperature resistant
Water resistance	Limited water resistant	Water resistant	Water resistant	Water resistant

The important properties of adhesive systems used in Germany most often, are summarized in table 1-2. Best properties concerning cohesiveness and adhesive strength are achieved by solvent-based adhesives and radiation-curing compounds. One important criteria for operation of industrial installations for the production of adhesive tapes is the maximum **coating velocity**. The coating velocity does not only depend on the construction of the coating installation but mostly also on the application weight. For example, acrylate dispersions can be coated with a sheeting velocity of more than 200 m / min if the application weight amounts only 20 g/ m^2. For application weights of 50 to 100 g/m^2, the coating velocity is reduced to 10 m/min due to drying capacity and drying kinetics (prevention of bubbles in the adhesive bulk).

1.2.2.2 Applications of Pressure-sensitive Adhesives

The selection of the adhesive system depends on the technical application of the adhesive tapes. Table 1-3 gives an overview over different applications of adhesive tapes and the adhesives most commonly used.

Table 1-3: *Applications of pressure-sensitive adhesives for adhesive tapes* [KREUTER 2001]

Utilization of Tape	**Application**	**Pressure-sensitive adhesive**
Packaging	Adhesive labels, adhesive tapes for packaging	SIS block polymer as hot melt, ***solvent-based natural rubber adhesives (SB),*** water based acrylate dispersions
Paper industry	Splicing of paper rolls, transfer adhesive tapes	Water soluble polyacrylates
Automotive industry	Transfer adhesive tapes for decorative strips and flank protection strips , insulating slabs, assembly aids for wheel case covers and side skirt pannels, adhesive tapes for winding of wire harnesses	***Solvent-based natural rubber adhesives, esters of acrylic acid and acrylates***
Construction industry	Foamed plastic adhesive tapes for sealing of windows, for caulking of construction splices and expansion joints, carpeting adhesive tapes etc	***Acrylate dispersions based on solvents*** or as watery dispersion
Medicine	Medical tapes, adhesive plasters	***Pure acrylates, polyacrylates***
Furniture industry	Transfer adhesive tapes for mirrors etc.	***Acrylate adhesives, solvent containing*** or watery

Solvent-containing adhesives are printed in italics

Packaging adhesive tapes, coating / masking adhesive tapes (including use as an assembly help) and double-sided adhesive tapes (transfer tapes) are produced in largest amounts.
For adhesive tapes produced in Europe, packaging adhesive tapes have a proportion of 74%, coating adhesive tapes only 10 %. Solvent-based adhesives have a proportion of 49 % in the European adhesive tape production. Hot melts have a proportion of 33 % and dispersions 18 % [LACOSTE 2001]. Table 1-4 summarizes adhesive systems used for these applications.

Table 1-4: *Types of pressure-sensitive adhesives and their applications* [RÖBER, WEIßLEDER 2001, HENDEß 1998]

Solvent	Acrylate	Natural caoutchouc	Synthetic caoutchouc
Dissolution in organic solvents	***Double-sided adhesive tapes***	***Packaging adhesive tapes, cover adhesive tapes***	***(negligible)***
Water based adhesive dispersions	Double-sided adhesive tapes and packaging adhesive tapes	(currently in development stage)	(***negligible***)
100% solids content, solvent-free	Double-sided adhesive tapes	(currently in development stage)	Packaging adhesive tapes, cover adhesive tapes, double-sided adhesive tapes

Product groups including solvent-containing adhesives are printed in italics

1.2.3 Production Process of Adhesive Tapes

The production process can be subdivided into three main steps:

- Surface treatment of substrate and coating of adhesive onto the sheet-like substrate (cf. fig 1-3)
- Drying, cross-linking and cooling (for hot melts) of the adhesive
- Coiling, cutting etc.

Figure 1-2 illustrates the production process of adhesive tapes with solvent-based adhesives. Exhaust gas is cleaned by condensation in pre-drying stage and by adsorption on activated carbon in the drying stage. Solvents are recaptured by steam desorption (cf. annex 5) and trapping.

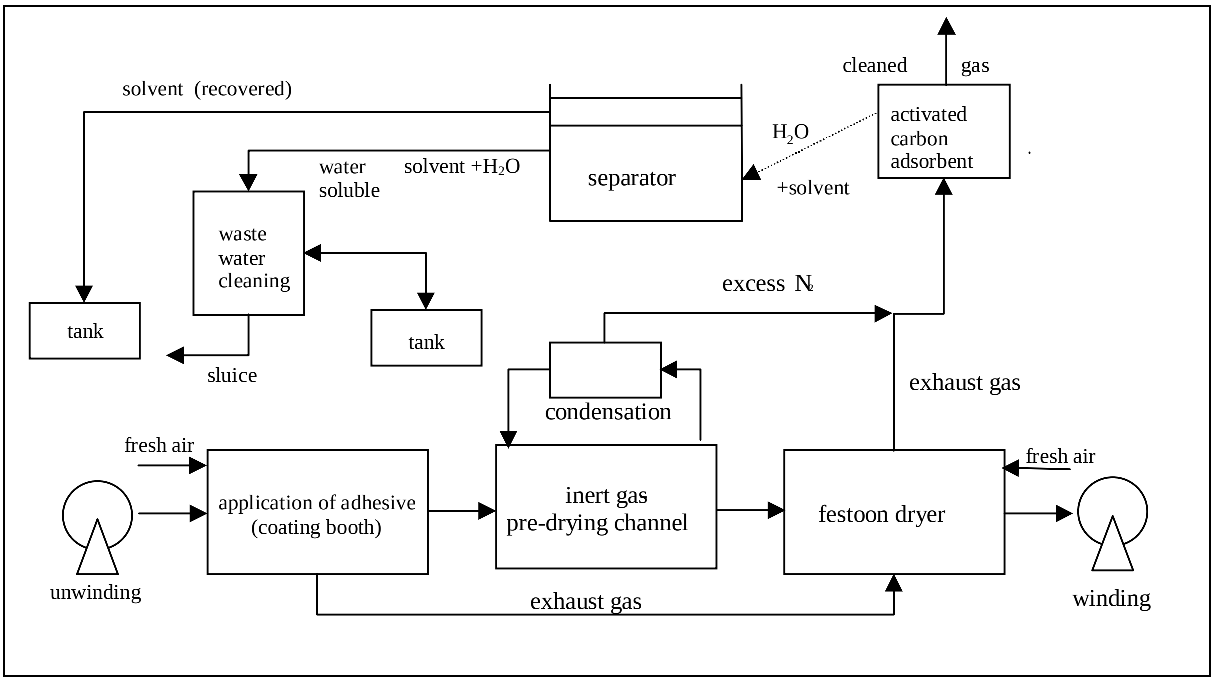

Figure 1-2: *Production of adhesive tapes with solvent-based adhesives and exhaust gas cleaning* [RÖBER, WEIßLEDER 2001]

In the following, the production steps of adhesive tape production with relevance for the environment will be discussed.

<u>Adhesive Coating</u>

The application of a pressure-sensitive-adhesive can be carried out by different application techniques. For processing of solvent-based adhesives doctor knife systems, accugravur- or reverse-roll-coat systems are used. For adhesive dispersions, doctor knife systems, reverse-roll-coat systems or roll doctor knife systems are appropriate [KREUTER 2001].
Hot melts are appliqued by doctor knife systems, slot die systems or extrusion techniques. Rarely, spraying techniques are used. Excessive material or overspray is coated by a doctor knive and re-used [HENDEß 1998]. The adhesive material is appliqued onto the substrate or a releasing material. For the so-called transfer technique, the pressure-sensitive-adhesive is dried onto the releasing material and cross-linked. Afterwards, the adhesive film is transferred onto the substrate by lamination. Therefore, the substrate is protected from damage during the drying process [KREUTER, RÖBER, WEIßLEDER 2001].

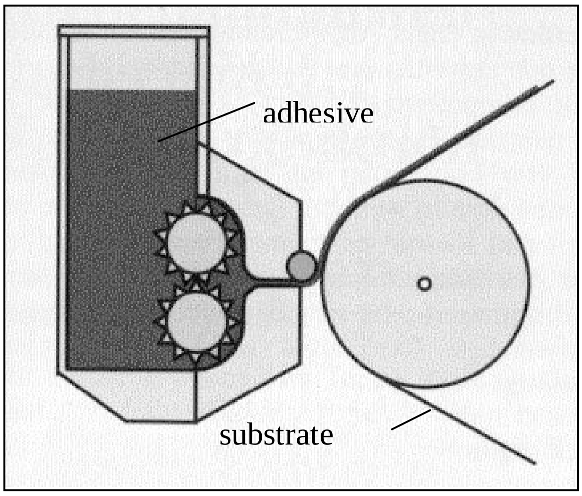

Figure 1-3: *Application of hot melts by the roll doctor-knife technique* [HENDEß 1998]

Drying

Air circulation dryers, infrared dryers and radiation systems (UV radiation or electron beams for cross-linking only) are predominantly used in industrial production. Solvent-based adhesives and dispersions are dried thermal (convection dryers and infrared dryers). Afterwards the cross-linking can be carried out thermally, chemically or physically by radiation in order to improve technical properties.

Hot melts harden autonomous during the cooling process. If solvent-based adhesives are used, solvent recovery installations are generally located downstream from the dryers.

- **Convection dryers** are used for dispersions and solvent-based adhesives. The air of these dryers is heated with gas or steam via heat exchangers or fuel oil. Besides the evaporation process of water or solvents, cross-linking steps take place in the adhesive material.
  If water based dispersions are dried, modern installations do not need more than 4000 kJ per evaporated kg of water (energy losses and heating of the substrates included) [KREUTER 2000]. When drying solvent-based adhesives the air is allowed to contain a certain maximum solvent concentration. The calculation of this maximum concentration follows EN 1539 and regulates a limit value of 50% of the lower explosion limit (UEG[1]). A reduction of the solvent concentration of the dryer's air is controlled by the air flow-rate. If gas-tight inert gas dryers are used, loads of 1200 g per m^3 nitrogen and more are possible. The load depends on the used solvents and the drying temperature [RÖBER, WEIßLEDER 2001].

[1] UEG: Lower Explosion Limit

- In **infrared dryers**, the medium to be dried is heated by infrared radiation. The emission spectrum has to be adjusted to the absorption spectrum of the material being dried. The technique is suitable for dispersions and also for solvent-based adhesives if explosion protection is considered.

Radiation Curing

- Radiation curing is used increasingly in the production of adhesive tapes. Curing by UV radiation and electron beams are the applied techniques. Due to lower investment needs, the UV radiation curing is predominant. UV radiation curing is especially applied for newer acrylate hot melts. The technique is suitable for radiation-curing adhesives. In these adhesives, the final reticulation and therefore adjustment of technically required cohesiveness and adhesive strength is initiated by UV radiation. For this process, glow-discharge lamps of high or middle pressure mercury vapour are used for a wavelength range of 180 to 300 nm. The ozone created by the radiation is extracted and leaded to a post-combustion installation. The installation has to be equipped with UV radiation protection. The radiation dose of common adhesives is in the range of 100 to 150 mJ/cm^2 [KREUTER 2001]. Advantages of the method are the low investment needs for the installation, the small space requirements, a high line speed as well as the possibility to retrofit existing installations [KREUTER 2001].

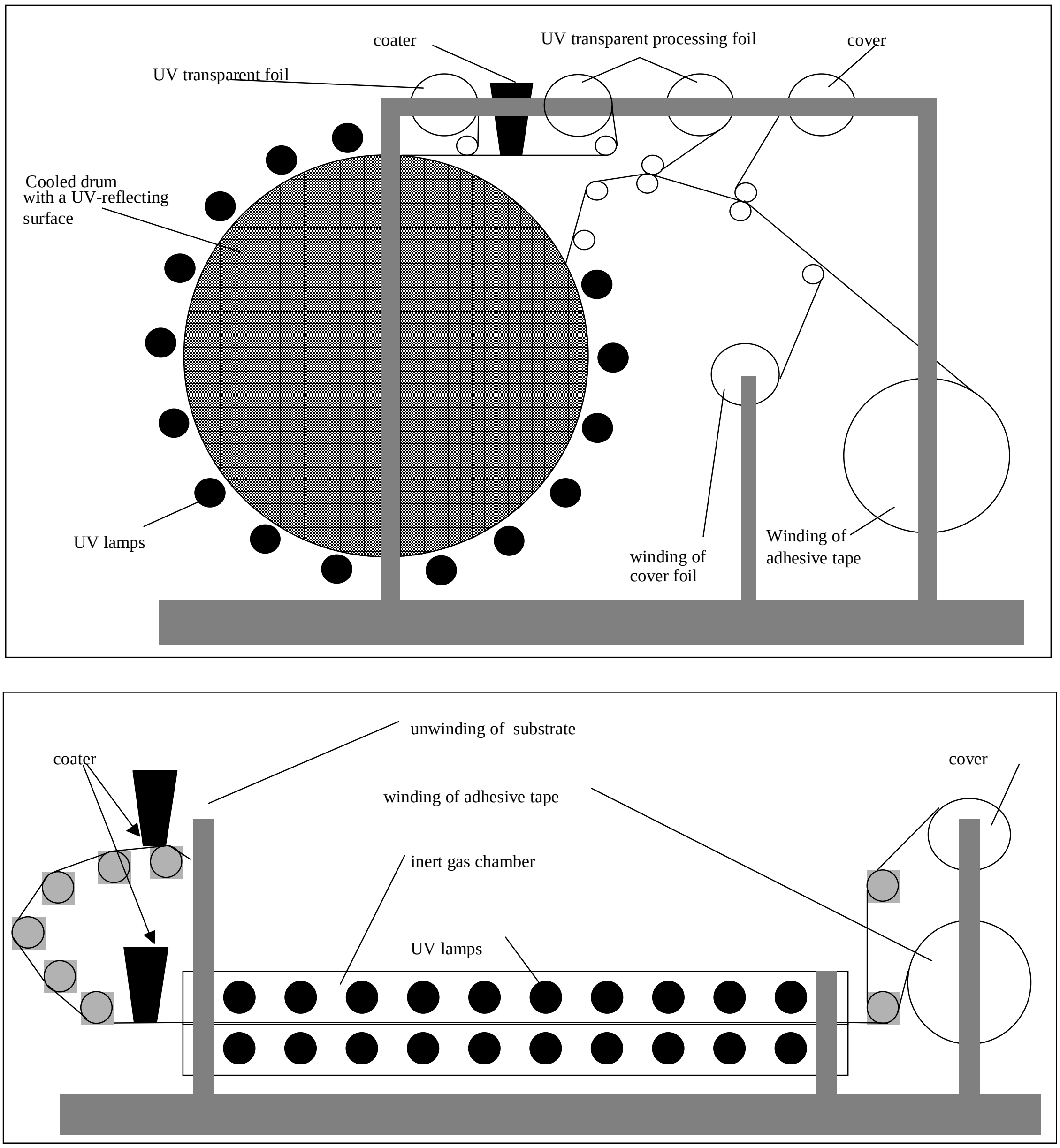

Figure 1-4: *Installations for production of one-sided and double-sided adhesives tapes (bottom) with UV cross-linking pressure-sensitive adhesives according to the transfer technique* [ASTORPLAST 1999]

- For **curing via electron beams**, the coated material is irradiated with electron accelerators. The created beams of 150 to 300 kV have to be shielded by lead plates or concrete walls. The electrons emitted by the electron accelerator are slowed down in the adhesive layer. The emitted energy initiates the cross-linking reaction. Depending on the cross-linking density, line speeds of up to 600 m/min can be achieved. The cross-linking by accelerated electrons has the advantage that the cross-linking is independent of the layer thickness. Therefore also adhesives that are not transparent (e.g. due to a higher proportion of filling agents) can be hardened reliable. A disadvantage compared to the UV curing is that changes in properties of the substrate are possible [KREUTER 2001].

Treatment of VOCs emitted in Drying Processes of Solvent Adhesives

Drying followed by solvent recovery via condensation

If a convection dryer is used as an inert gas dryer, big amounts of solvents can be evaporated and recovered. For the inert gas drying process, a gas volume of 2000 m³ is circulated for a solvent amount of 400 kg/h. However if air drying is used, up to 20.000 m³/h are necessary to meet the requirements of 40% of the lower explosion limit (UEG[2]). (This means up to the tenfold of the gas volume of a conventional air dryer) [RÖBER, WEIßLEDER 2001].

For recovery, the gas-solvent mixture is cooled down (5 to –30°C) by a cooling system until the dew point of the solvent is reached. The condensed solvent is leaded into tanks . The dried gas is re-heated and leaded back to the dryers gas supply continuously. The recovered solvents can be utilized again for the process of adhesive production [RÖBER, WEIßLEDER 2001]. For example, the technique is used in the pre-drying step (cf. annex IV volume application of varnishes).

Drying followed by solvent recovery by sorption on activated carbon

The exhaust gas loaded with VOCs flows through adsorbers that are generally constructed as several parallel connected tanks. The tanks are filled with activated carbon (cf. annex V, volume paint application). If one absorber is saturated with VOCs, the VOC loaded exhaust gas is diverted to the adjacent adsorber. This allows a recovery of intake capacity of the adsorption agent of the saturated tank by regeneration. For regeneration, the adsorbed VOCs are first desorbed due to a rise in temperature initiated by steam supply. Then the water-solvent mixture being formed is condensed and parted by phase separation [RÖBER, WEIßLEDER 2001]. This technique submits VOC concentrations in the cleaned gas below 100 mg/m³ and therefore meets the requirements for Technical Instructions on Air Quality Control for installations with solvent recovery. With the current mode of operation, concentrations of 70 to 90 mg/m^3 are achieved in the cleaned gas. Solvents recovered by the desorption can by supplied again for the production process and especially for the production of solvent-based adhesive bulks. About 2 to 3 tons of steam are necessary per ton desorbed solvent in order to achieve a cleaned gas concentration of 70 to 90 mg/m^3 [RÖBER, WEIßLEDER 2001]. Lower concentrations in the cleaned gas can be achieved by a reduction in adsorption periods and are hence correlated to an increased steam consumption.

[2] UEG is the German abbreviation for Lower Explosion Limit

1.3 Current Consumption and Emission Values

Partly, more than 2000 t/a solvent-based adhesives are processed in large industrial installations of adhesive tape production. The emission limits for installations of adhesive tape production set by 31. BImSchV are undershoot significant due to the use of exhaust gas cleaning techniques. For example the condensation method is used in pre-drying and the adsorption method in the drying step (cf. Table 1-5).

Table 1-5: *Limit values for fugitive and untreated emissions in waste gases according to 31. BImSchV*

Solvent consumption [t / a]	**Limit value for fugitive emissions for new installations [% of used solvents]**	**Limit value for emissions in untreated waste gases [mg C/m³]**	**Comment**
> 15	20	50 100 (1)	For automated coating of sheet-like materials

(1) if solvent recovery techniques are used, if a thermal exhaust gas cleaning is used 20 mg C/m³

Adhesive tape production with solvent-based adhesives was studied at the Beiersdorf AG, Hamburg in the installation Hausbruch (cf. case study annex A). The solvent consumption of the investigated installation amounted 2350 tons in 1999. Emissions of VOCs arise during application of solvent-based adhesives in the following areas:

- Remaining solvent in the adhesive tape ready for use:
  Remaining solvent proportions of finished double-sided adhesive tapes produced with solvent-based adhesives account for between 0.1 and 2 % based on the used amount of solvent. Solvents remaining in the products are either slowly lost due to diffusion processes or they remain in the adhesive layer functioning as a plasticizer [RÖBER 2001].
- Coating
- Drying
- Mixture of components in order to produce an adhesive ready for use
- Cleaning of machines (coaters)
- Emissions in the wastewater from adsorption techniques
- Adhesive waste

0, 85 % of the used mass of solvents is emitted, 5,8% remain in the adhesive, waste and wastewater and are recycled (material recycled/or thermally). 93,35 % of the used solvents are recovered. The recovered solvents are mostly recycled in the production process. Remaining amounts that can not be reused due to quality reasons are sold as regenerative solvents substantial recycling. Water-soluble solvents that have to be removed from the wastewater of the adsorption installations (e.g. by rectification) can be used for heating purposes or they are recycled by distillation [RÖBER, WEIßLEDER 2001].

1.4 Determination of the BAT Candidates

Dispersions, UV cross-linking adhesives and hot melts distinguish themselves positively concerning solvent emissions and energy consumption compared to solvent-based adhesives, because for these systems, no energy-rich exhaust cleaning measures have to be carried out or only to a very low extent.

Hot melts have a poor resistance against aging and a low UV stability and show a thermoplastic behavior and high adhesion while their cohesiveness is low. For these reasons, this adhesive system is suitable for coating- and packaging adhesive tapes of the lower region of the quality range. Water based dispersions show better cohesiveness but have a very limited water resistance. UV cross-linking acrylates and electron beam curing systems do not show the disadvantages of the hot melts and dispersions and are distinguished by high cohesiveness and high adhesion strength. However these systems cannot be used for all products yet, because the cohesiveness and adhesive strength that can be achieved with solvent-based adhesives can not be reached due to shorter polymer chains. Therefore, no alternative to the solvent-based acrylate and caoutchouc adhesives does exist for very durable or mechanical and thermal high stressed adhesive bondings. If solvent-based adhesives are used, VOC emissions can be avoided efficiently by exhaust gas treatment measures such as adsorption and condensation.

1.5 Proposal for the Best Available Techniques

Adhesive tapes are produced for various purposes and therefore show very different technological properties. The used adhesive system depends on the coated product and its required technical properties. Hence the selection of one uniform BAT for the production of all the sector’s various products is not possible, but has to be oriented on the required properties of the adhesive:

- Hot melts and dispersion for the lower quality range of packaging and masking tapes as well as double-sided tapes.
- UV cross-linking systems for solvent-based adhesives up to the lower quality range of transfer and packaging tapes as well as masking tapes.
- Solvent-based adhesive bulks for adhesive tape applications that require a high stress resistance. Up to now, there are no solvent-free adhesives available that show equivalent properties. Therefore, there is no alternative to solvent-based adhesives in

the high quality range. For these applications, techniques such as adsorption installations and inert gas dryers followed by condensation installations are available that reduce emissions to the environment and allow a re-use of solvents. The emission values of the 31.BImschV are achieved.

1.6 New Progressive Techniques

UV- cross-linking pressure-sensitive-adhesives and dispersions based on caoutchouc for high-resistant adhesives are currently developped [RÖBER, WEIßLEDER 2001].

1.7 Conclusions and Recommendations

A far reaching substitution of solvent-based adhesives has been observed in big partitions of the sector. For some partitions, avoidance of solvent-based adhesives is not possible yet due to the specific properties. As there are no solvent-free products with equivalent properties available, subsequent techniques are used for reduction of VOC emissions and recovery of solvents. It is estimated that the proportion of solvent-based adhesives will decrease with further development of UV cross-linking adhesives or dispersions.

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2 Paper and Packaging Industry

Among the total consumption of adhesives in the paper and packaging industry, dispersions have a share of 50 %, hot melts of 23 % and adhesives based on natural polymers have a proportion of 21 %. Solvent containing adhesives are used with a share of 6 % in the paper and packaging industry [BMU 1997]. According to statements of consulted experts and manufacturers, solvent containing adhesives are processed in the production of composite foils exclusively [HVP 2000, FKN 2000]. Therefore only this application is considered in the following.

2.1 Production of Compound Foils

2.1.1 General Information

The threshold values for the consumption capacity of organic solvents specified by the Directive (annex I, 6.7) are exceeded by a multiple in the production of composite foils depending on the applied technique. In the lamination of composite foils, a combined material is created out of two or more foils. This is further processed to packages for example. Due to the construction of composite foils, a combination of different properties of the single components is achieved. A property profile is created that is focused on the corresponding application purpose. The individual foils have to meet different requirements such as printability, aroma barrier, moisture barrier or mechanical toughness.

Composite foils are processed to so-called flexible packages (e.g. snack packages), packages for pet food, packages for chemical products (e.g. pesticides, fertilizer, detergents), packages for metal products, (e.g. replacement parts, stamped parts), packages for medical and pharmaceutical products (e.g. pills, vitamin powder), packages for tiles (e.g. building materials, substrates) and sanitary products [WIEGEL 2001]. With respect to adhesive application there are significant differences between consumer- and industrial packages.

Industrial packagings are packages (e.g. bags, sacs) for larger amounts of content. Industrial packages typically consist of mono materials or the different materials are cemented only selectively in seam and edge areas. Materials cemented completely are manufactured for production of composite foils almost exclusively. Production of composite foils for food packaging has the highest proportion in the lamination of composite foils. Within this sector the production of snack food packages is of utmost significance [VDI 1512]. Due to small amounts of adhesives used in the production of industrial packages, only production of consumer packages will be discussed in the following.

In 1999, 330.400 tons of multiple layer foils with a value of 1.045 million EUR were produced in Germany [IKV 2000]. 536 enterprises with 72.952 employees worked in the production of plastic foils in 1999 [STATISTISCHES BUNDESAMT 1999].
A strong increase in the use of solvent-free adhesives has been observed in the production of composite foils. Whereas in 1975, solvent containing adhesives were used in lamination exclusively, the share of solvent-free products was already 40 % in 1999 [HENKE 2000].
The total adhesive mass processed for production of composite foils in Germany in 1999 amounted 5.000 to 10.000 tons [FRICKE 2000]. The production of composite foils has been studied in two plants.

2.1.2 Applied Processes and Techniques

Co-extrusion techniques, extrusion coating techniques and lamination techniques are used for production of composite foils. With respect to the use of adhesives, lamination is relevant. Laminating is a process where two or more different plane materials are permanently joined to a composite over the complete surface area [VDI 2000].

2.1.2.1 Composite Materials for Foil Lamination

About 60 to 70 different foil materials are used for the production of composite foils. The choice of an individual foil and an adhesive system limited by the material being packaged, by the packaging machine as well as by specific product requirements [SCHMID 1997].

Hence the sensitivity of the material being packaged against light, oxygen, moisture and aroma loss determines the barrier properties of the composite foil. The desired stability period determines the quality of the required protection. Often food packages are filled with protection gases in order to prevent spoilage and oxidation of the food.
The foil has to be gas tight during the storage period of the product in order to maintain the artificial atmosphere within the package.
Chemically aggressive substances contained in the material being packaged such as oils, fats and alcohols determine the required resistance of the material against these substances.

Further also the packaging machine determines requirements of the composite. Critical parameters for processing capability of a material are for example mechanical stability, gliding properties or flatness. Here the sealable coating has a high significance, because its properties determine the processing velocity of the foil.

Depending on the application purpose of the foil, additional requirements have to be met, such as cutting toughness, tensile strength, kink-resistance, fracture resistance, temperature resistance (pasteurization, boiling, sterilization), printability and mechanical stability.

Lamination adhesives have to meet different requirements. For example physiological harmlessness, machineability, starting and final adhesive strength, wettability as well as cross-linking velocity are critical parameters in the selection of lamination adhesives for production of composite foils for food packages.
Besides these factors, the selection of adhesives is also influenced by requirements for the finished packaging material. These are requirements such as the behaviour during the packaging process, mechanical properties of the package, barrier and content resistivity durability, odour neutrality, taste neutrality and heat resistance [VOSS 1993]. For printed foils, the compatibility of the adhesive system and printing ink has to be considered because adhesive and printing ink often have a direct contact. A good penetration of the printing ink by the adhesive has to be guaranteed for high composite strength values.

Composite foils can be classified as plastic and aluminum foils. For example polyethylene, polypropylene, polyamid and ethylen vinyl alcohol are processed for **plastic foils**. Foils may be stretched mono-axially or bi-axially or metallized in order to achieve specific properties.

Composite materials for **aluminium foils** are plastic foils, allophane, paper, cardboard or a combination of these materials. Aluminium serves as a barrier against gases, steam, aromas and light.

2.1.2.2 Lamination Techniques

Presently, solvent-based adhesives are used only partly in the so-called wet and dry lamination. In the following, these techniques will be discussed. Adhesive application is carried out via coaters. Generally these are encapsulated and equipped with a extraction installations in order to ensure an undershooting of the respective limit values (the lower explosion protection limit and the maximum work place concentration) [VDI 2587/3].

The adhesive is dried before bonding of the materials if the **dry lamination technique** is used (cf. figure 2-1). In the **wet lamination technique**, materials are bonded together immediately after application of the adhesive. For both variations, solvent-free adhesives are processed increasingly. The bonding of the two materials is carried out between lamination rolls, lamination drums or in a press under temperature and pressure. The material is supplied from coils.

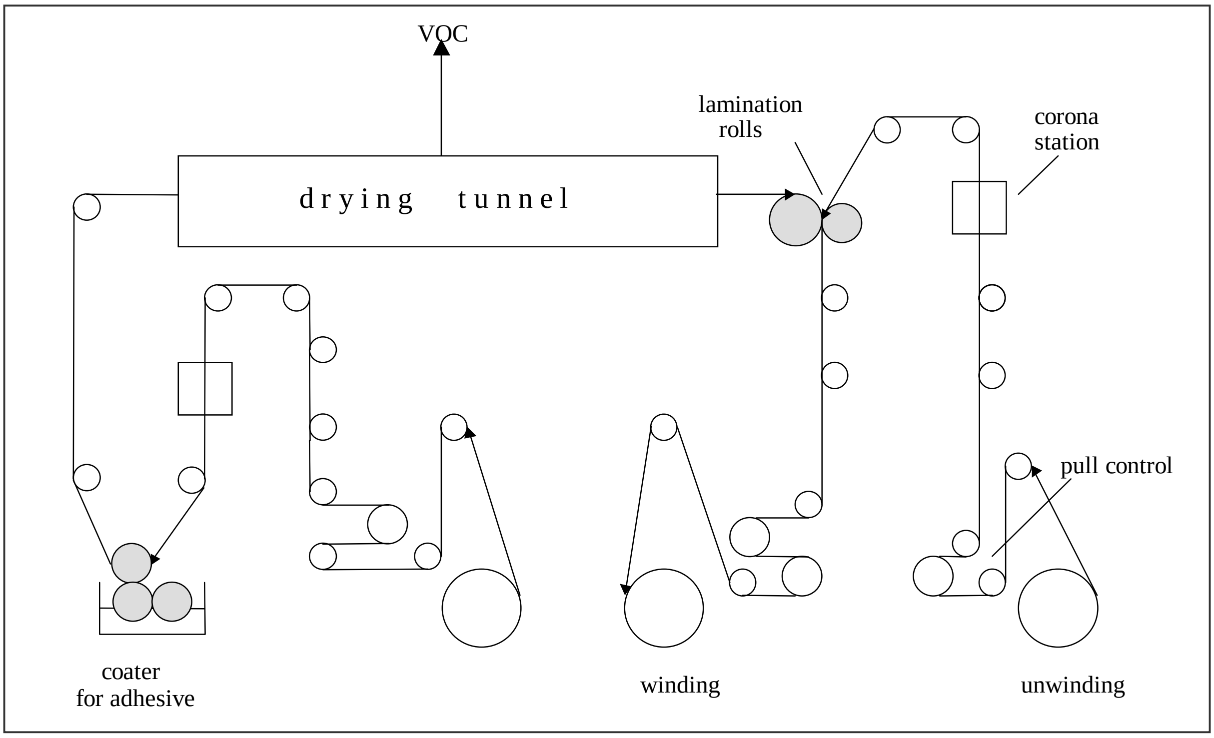

Figure 2-1: *Dry lamination technique with solvent-containing adhesives*

In the **dry lamination** technique, the adhesive is applied onto a substrate and dried afterwards (cf. Figure 2-1).The application rolls are constructed as plain or grid rolls. The drying tunnel consists of different chambers, where the temperature can be adjusted independently. If solvent-based adhesives are used for lamination of e.g. paper and metallized foil, typically amounts ranging between 2,5 and 4,5 g/m², are appliqued. After drying, the substrate coated with the adhesive is merged with the second substrate in the lamination gap. In general, lamination rolls are heatable. The produced composite is winded afterwards.

Solvent-free adhesives can be used for all common films and aluminum foils in dry lamination. Limitations of the solvent –free adhesives are the lower adhesive masses that can be appliqued. Hence these adhesive systems are not suitable for multi-layer films for production of packages that are going to be filled with aggressive substances or that have to resist extreme sterilization conditions. Adjustment of adhesive viscosity may be regulated by temperature adjustment of the application rolls for hot applicable adhesives.

Water based adhesive dispersions based on dextrin, casein or solvent-based adhesives are appliqued in **wet lamination**. Aluminum foils and paper substrates are laminated. In the laminating process, the waterproof substrate is coated with the adhesive. The water is absorbed due to the porosity of the other substrate. For example paper is placed onto the adhesive coated aluminum foil (Fig 2-2). The wet lamination method is used for cigaretts, tea, chewing gum and soap final packages [VDI 2000].

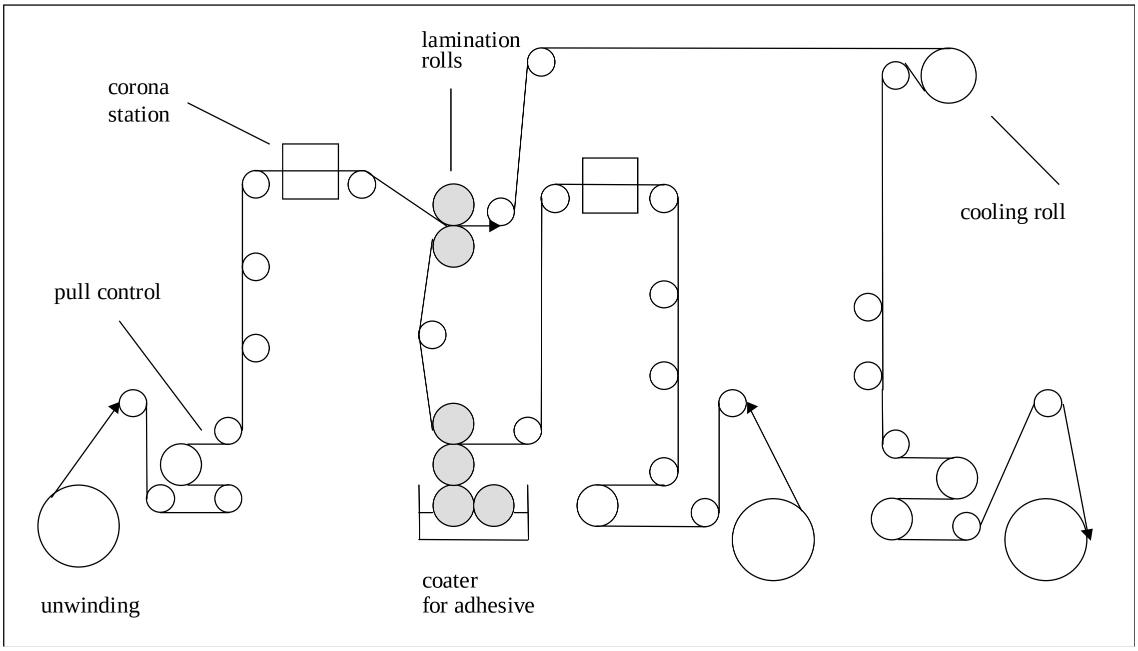

Figure 2-2: *Solvent-free wet lamination technique*

Typically, amounts of 1 to 2 g/m² (oven dry mass) are reached by wet lamination with solvent-free adhesives. For composites with paper, the appliqued amount may be higher due to the absorbancy of the material.

In the solvent-free lamination, there is a low beginning adhesive strength in the composite. This results in problems in laminating printed foils together with lower adhesive masses being applied. Especially large surface printed foils, foils coated with the locking printing technique or other surface irregularities originating from printing of different ink layers may result in voids and de-lamination. Further, solvent containing printing inks have to have cured completely before any application of adhesives [SCHMID 1999].

2.1.2.3 Surface Preparation before Lamination

Polyethylene and Polyolefin foils posses a non-polar, electrically well isolating and hydrophobic surface. This surface is poorly wettable and needs to be prepared before coating with adhesives or printing inks. Preparation techniques are corona or flame pretreatment or plasma treatment (to avoid de-lamination after bonding). The preparation method depends on the utilized substrate. For the pre-treatment no solvents are used..

2.1.2.4 Printing Processes

The materials being bonded (foils, paper, cardboard) are often printed before or after the lamination by the flexo-offset printing and gravure techniques. Printing is seldom carried out in combined installations (inline operation). As solvent-based adhesives as well as solvent-based printing inks are used, VOC emissions cannot be associated with lamination or printing clearly. In newer installations, the printing and lamination installations are constructed separately with an increasing tendency. Reasons are the different sheeting velocities of the installations as well as the reduced susceptibility to faults of the installations [KAHL, WIEGEL 2001]. Solvents (ethanol, ester) are used for cleaning of lamination rolls and printing mechanisms. However in general, a separation of the used cleaning solvents with respect to the use for printing mechanisms or lamination installations is not possible [KAHL 2001].

In 1999, proportions of the different printing techniques for flexible packages amounted to 16 % for the offset printing, 50% for gravure and 34 % for flexo printing [WALMAN 1999]. In the production of composites for consumer packages, several hundred different inks are used [RHEINISCH 2001]. For package printing of composite foils, nitrocellulose inks (NC) are used predominantly. Further, polyvinyl butyral (PVB) and polyvinyl chloride (PVC) are applied. The printing inks typically consist of 65 – 80 % solvent, 20 % pigment binder and 12 to 16 % pigments. As organic solvents, alcohols, esters and ketones are used [MEYER-ROSCHER 1999]. Partly also water-based printing inks are processed.

Some individual foils are coated with sealing layers additionally. The solvent-based products that were used in the past have already been replaced by dispersions in new installations [NETWIG 2000]. Generally the dryer is operated under low pressure if solvent-containing inks are used and the exhaust gas is leaded to a waste gas treatment installation. Treatment of exhaust gas can be carried out by thermal or catalytic combustion or by bio-filtration techniques (see also annex V in volume varnish application, also the VOC can be recovered [VDI 2587].[1]

[1] Depending on the applied technique, concentrations of about 10 to 20 mg C/m3 can be reached in the clean gas by thermal or catalytic exhaust gas cleaning. More detailed information can be found in [VDI 2587].

2.1.2.5 Adhesives for Lamination Techniques

Lamination adhesives are typically appliqued as liquid phase. In the production of composite foils, application of solvent-based adhesives were predominant until some years ago. Presently these components have been replaced prevalently by solvent-free systems in the mass production of composite foils.

The binding agents exist here either dissolved in an organic solvent or dispersed in water. Among the newer developments there are high solid systems having a solids content of more than 60 % [NETWIG 2000].

Adhesives used in the production of composite foils are classified in the branch according to their performance range [VDI 2000]:

General-Purpose Adhesives

General-purpose adhesives are used for lamination of transparent and metallized substrates as they are used for packaging of dry, chemically non-aggressive materials.

Medium Performance Adhesives, Multi Purpose Adhesives

Medium performance adhesives are processed in the lamination of substrates that are used for production of packages for paste-like or liquid, fatty or slightly chemically aggressive materials. Due to the development of adhesives based on polyether or polyester polyurethane it is possible to combine adhesives for a variety of substrates (multi purpose adhesives).

High Performance Adhesives

High performance adhesives are used for lamination of substrates that have to meet high requirements concerning product stability and composite durability. The applied adhesives have to prevent fluid or chemically aggressive materials from migration or they have to resist sterilization conditions as well as to show a high adhesion strength on aluminum. During the sterilization process, the materials have to resist temperatures of up to 139 °C. Hence these adhesives are used in the production of composite foils for medical products. As fig 2-3 illustrates, the highest shift towards solvent-free systems has taken place for the general-purpose adhesives. Also for medium-performance adhesives, the proportion of solvent-free adhesives has increased strongly. However, only low substitution has taken place in the group of high-performance adhesives.

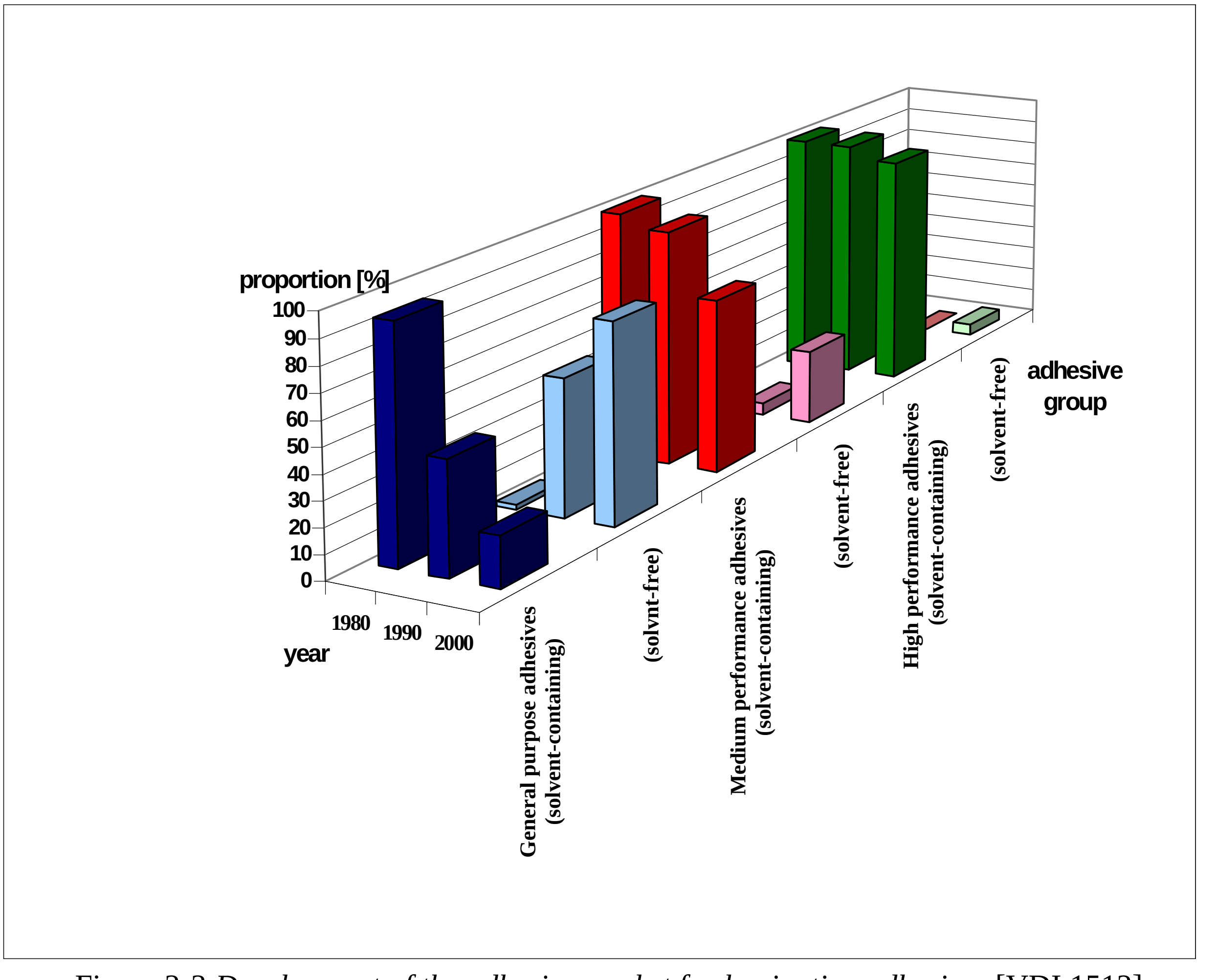

Figure 2-3:*Development of the adhesive market for lamination adhesives* [VDI 1512]

<u>Adhesives with organic Solvents</u>

Predominantly there are one- or two part solvent-based adhesives. Application masses of 2 - 6 g/m^2 (dry) are obtained. Predominantly the following two systems are used [VDI 2587/3]:

- *Physically drying adhesives*: E.g. nitrocellulose adhesives with a solvent proportion of 35 to 75 % (weight per cent).
- *Reactive adhesives*: E.g. isocyanate cross-linking polyurethanes or epoxy resins with a solvent proportion of 20 to 40 % (weight per cent).

Typical solvents are hydrocarbons, alcohols, esters and ketones. Advantages of solvent-based systems are the high starting adhesive strength, the high composite adhesion and the resistance against chemically aggressive media. Viscosity of the adhesive and the application mass can simply be regulated by the solvent content. With this technique, chemical resistant and sterilization resistant packages can be produced.

Partly, metal complexes are added to the adhesive for coloring purposes. These compounds are soluble in organic solvents only. Hence they can be processed in solvent-based adhesives exclusively. For example this technique is used for production of foils for coffee packages.

As lamination substrates, all common films and aluminum foils can be processed. Hence solvent-based adhesives are used especially for processing of small and fast changing adhesive spreads or for production of foils designed for high thermal and mechanical stress. Application of just one adhesive system that is suitable for a variety of composites does reduce the change-over times following a change in material and does strongly decrease the amount of waste. If solvent-based adhesives are used, an exhaust gas cleaning is necessary [VDI 2587/3].

Adhesives with Water as Dispersing Agent and Solvent

- *Adhesives drying physically*:

-Acrylic dispersions

-Polyvinyl acetate dispersions

-Ethylene vinyl acetate dispersions

-latex dispersions

-PVC dispersions

-polyurethane dispersions

- *Reactive cross-linking adhesives:*

-isocyanate cross-linking water-soluble polyester polyols

-epoxy cross-linking watery polyurethane emulsions

Partly watery dispersions still contain low proportions or organic solvents. If water based printing inks are used, reactions between ink and adhesive may take place and wetting problems may arise [VOSS 1993].

For an increase in drying velocity, adhesives soluble in water alcohol mixtures may be processed. For these systems, the drying time can be reduced drastically compared to water as solvent [VOSS 1993].

If water is used as a solvent or dispersing agent, the substrates being bonded have to possess at least partly an absorbent surface so that water can evaporate from the adhesive layer. Hence only combinations of foil or aluminium with paper can be produced with water dilutable adhesives. A further problem of these adhesives is that sufficient drying of the adhesive layers is only ensured for application weights up to 2 g/m². Generally, the energy demand for physical drying is higher than for the drying of solvent-based adhesives.

Adhesives without any Solvents (organic or watery)

- *hot melts*
- *reactive cross-linking adhesives*
  (one and two part radiation-curing adhesives, systems with 100% solids content often contain isocyanates)

A general problem of solvent-free so-called 100% systems is the application of thin, uniform adhesive films. As opposed to the solvent-based adhesives where layer thickness can be regulated by the viscosity (solvent content), the regulation has to be carried out by mechanical rolling processes. The adhesive layer shows slightly increased surface irregularities compared to solvent-based adhesives.
The solvent-free adhesives for lamination of the 1st generation were moisture curing one part polyurea polyurethane adhesives. For these adhesive systems, a complete curing when bonding foil or aluminium cannot always be ensured even if steam supply from outside is provided. Hence curing faults may arise. Therefore the adhesives are applied for bonding of paper composites, where material moisture supports curing. For these materials the reduced adhesion strength compared to solvent-based systems is of minor significance. Further, the adhesives still contain remaining monomers after cross-linking. These can enter the material being packaged by migration [HENKE 1999].

Solvent-free adhesives of the 2nd generation are two part systems based on polyester with polyisocyanate as curing agent. The curing process can be better influenced than with moisture curing one part adhesives by the dose of curing agent. Adhesives are liquid and can be applied at a temperature in between 25 - 45° C. Polyesters show a better adhesion on metallic surfaces and hence they can be used for aluminium foils or aluminium vapour coated foils. These adhesives are also characterised by the low starting strength. Especially on gliding agent containing foils, on ethylene vinyl acetate and white-pigmented polyethylene, anti-sealing effects may arise.

The 3rd generation of solvent-free adhesives is based on two part polyurethanes. In these systems, no free monomers are created during the curing process due to a complete reaction. Hence a migration of the monomers into the material being packaged is excluded. Adhesives can be used for all materials. Due to the high temperature resistance, packages can be sterilized by heat [HENKE 1993]. Mostly these adhesives are processed in lamination installations with a 3 or 4 roll coater specifically constructed for the adhesive material.
For solvent-free lamination, the sheeting tension of the substrate and the supply foil have to be adjustable precisely due to the low adhesive starting strength in order to avoid formation of crimps and curls in the composite. Sheeting velocities of 200 m/min can be achieved. The disadvantage of the solvent-free systems of the 3rd generation is their low curing. Curing of the adhesive takes about one week. This process can be reduced by 2 to 3 days maximum by tempering [HENKE 1995].

The 4th generation of solvent-free UV curing or electron beam curing adhesives is based on (one part) epoxies. Their advantage is fast curing, initiated by the irradiation. Thus a high bonding strength is achieved. The material can therefore be further processed without additional drying time [HENKE 1995]. So far these adhesives have only been used in the plastic sector or in the aluminium sector for standard composites up to a medium quality level.

2.1.3 Current Consumption and Emission Values

In general, exhaust gases of solvent processing dry lamination installations are leaded to an exhaust gas treatment installation, where the VOCs are utilised energetically via combustion or collected and re-used. Inert gas dryers (cf. chapter 1) being able to support an economic incineration of exhaust gases can not be used in the branch due to the frequent changes in material associated with frequent start up and shut down processes of the installations.
For the production of composites for consumer packages, solvent-based adhesives are the most used adhesives due to frequent change in materials, especially because they are able to bond different materials. In the investigated installation approximately 524 t of organic solvents were processed. From this amount approximately 392 t were recovered by absorption.

Emissions in the Air
From the investigated installation approximately 3,65 t were emitted via cleaned gas from the absorption installations.

2.1.4 Determination of the Best Available Technique Candidates

Integrated Measures

If solvent-based adhesives are used, high-solid adhesive systems with solids content of up to 60 % can make an effective contribution towards the reduction of solvent application.

Solvent-free adhesive systems are already prevalently in use for general purpose and medium performance adhesives applied for mass production of composites and standard composites of up to the highest quality segment. They represent an established BAT for this product segment. As these adhesives show a lower chemical resistance compared to solvent-based adhesives besides the lower adhesive starting- and composite strength, they cannot be processed for all types of applications up to now. Water based adhesives are only suitable for bonding of materials with absorbent surface. In this application, only applied masses of up to 2 g / m² can be applied in order to guarantee a complete drying of the adhesive layer. The produced composite has to be stored for some days in order to cure the adhesive layer before it can be further processed into packages.

For **solvent-free UV or electron beam curing adhesive systems** and **hot melts**, the adjustment of layer thickness has to be realised by mechanical rolling processes as opposed to dispersions and solvent-based adhesives. Hence layer thickness is more irregular.

Substitution of solvent-based polyurethane adhesives is especially carried out because solvents do not evaporate completely during the processing so that partly traces of solvents originating from the foil composite may be able to enter the food by diffusion processes. These adhesives were substituted by solvent-free, chemically curing adhesives such as watery polyurethane dispersions or watery polyacrylate dispersions [KAHL 2001, ADHÄSION 2000]. These two part systems are applied by a wet lamination technique with 3 and 4 roll-coaters. Adhesive application masses amount up to 1,6 – 2,1 g /m². Products are manufactured with sheeting velocities of 150 - 250 m/min [KAHL 2001]. Cleaning solvents for cleaning of roll-coaters and printing mechanisms can be re-used via distillation. Solvents are only used in the printing process besides cleaning of the lamination rolls [KAHL 2001].

Solvent-based adhesives allow bonding of all materials even with high applications of adhesive mass. Hence generally solvent-based high performance adhesives are used if the materials to bond change often in the lamination installation. At presence there are no equivalent adhesive systems available for packages that are strongly stressed by heat (sterilization) or are mechanically or chemically stressed.

Subsequent Measures

For the application of solvent-based adhesives, effective subsequent exhaust cleaning techniques have been established that partly allow a solvent recovery (cf. annex V and [VDI 2587/3] for a detailed description of these techniques).

Solvents contained in the exhaust gas, can be recovered by **absorption onto organic oils** followed by distillation. This technique is only used for pure solvents, so-called mono-solvents, due to economic reasons. The recovered solvent can be supplied again for the production process. Presently there are no experiences with other solvents in Germany. The technique is used for waste gas loads with a minimum of 2 g/m^3. Concentrations of < 30 mg/m^3 are reached in the cleaned gas. Depending on the applied technique, about 0,1 to 0,5 m^3 wastewater are created per ton of solvent recovered [VDI 2587/3].

A further technique of emission reduction is the **adsorption** of solvents in the exhaust gas flow onto activated carbon and rarely onto zeolites. Installations with water steam and inert gas desorption are used. If there are more than two different solvents in the exhaust gas flow, no economic solvent recovery and recycling for the production process is possible any more. A substantial recycling as cleaning solution or a thermal recycling of the reclaimed material should be realized in this case. Increasingly also adsorber wheels are used. In this case, the desorption flow is leaded to a post-combustion unit. The technique is used for wastegas loads up to 4 g/m^3 Concentrations of < 20 mg/m^3 are achieved in the cleaned gas. The hot steam desorption method generates about 4 - 6 m^3 wastewater per mg of recovered solvent.

If changing solvent mixtures are applied with higher loads or after concentration, generally a **post-combustion** method is used. Exhaust gas flows with lower loads can be treated with a catalytic post-combustion unit (regenerative or recuperative) as well as with the thermal regenerative post-combustion units.

Cleaning solvents for cleaning of application rolls and printing mechanisms may be recovered by distillation installations leading to a reduction of the amount of hazardous waste that has to be disposed of. Solvents originating from adhesive leftovers can be recovered by distillation and be recycled substantially or thermally. Fugitive emissions can be reduced this way [RHEINISCH 2001].

2.1.5 Proposal for the Best Available Techniques

Due to the high variety of used materials and a similar construction of application devices, the selection of best available techniques is orientated best on the used adhesive system.

- For mass composites without stricter requirements the application of solvent-free adhesives is already an established BAT.
- Application of solvent-based adhesives with high solids contents (High Solid Systems) for the high performance sector for mechanically, thermally or chemically stressed packages and in the production of composites changing in material within one lamination installation. Subsequent exhaust gas cleaning techniques such as absorption and adsorption allow a high proportion of solvents to be re-used after distillation. For non-recyclable solvents (e.g. solvent mixtures), thermal or catalytic exhaust gas cleaning can be used (see also [VDI 2587/3]). The waste heat is generally utilised for the heating of dryers. Clean gas concentrations of less than 20 mg C/m^3 are achieved.
- Reduction of fugitive emissions by distillation of solvent containing adhesive leftovers and cleaning solvents followed by recycling of the solvents.

2.1.6 New Progressive Techniques

Possibly new developed solvent-free polyacrylate dispersions are suitable to substitute the solvent-based polyurethane adhesives for most applications in the snack food sector (general and medium purpose adhesives) [ADHÄSION 11/2000, MEYER-ROSCHER 1999].
Application of solvent-free adhesives is possible, because no extremely high adhesion strength is required for the composites of the snack food sector and additionally, food regulations by law may be fulfilled. It has to be considered that also the solvent-free adhesives (moisture curing or a dispersion) contain isocyanate if they substitute the solvent-based polyurethanes. (In the past, disadvantages of the acrylate dispersions have been their adhesion problems on nitrocellulose based printing inks). However the achievable peel strength of the acrylate dispersions of the newer generations is almost as high as the one of solvent-based polyurethane adhesives [MEYER-ROSCHER 1999].
Application of these solvent-free adhesive systems has been in a testing stage up to now [FRICKE 2000].

2.1.7 Conclusions and Recommendations

Application of solvent-based adhesives nowadays is limited to the so-called high performance sector. Powerful solvent-free adhesive systems are being developed presently for this sector so that a substitution of solvent-based adhesives is expected also within the highest quality segment.

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3 Vehicle Production

In this chapter, the adhesive application in the different production lines of the automotive industry will be described. Adhesives are used in the serial production of passenger cars, commercial vehicles, mobile homes and trailers as well as in the aviation and rail vehicle industries. Over the last years, a far-reaching substitution of solvent-based adhesives has taken place in automotive production processes. Installations whose consumption capacity exceeds the threshold values of the Directive (annex I; no. 6.7) are not known. The highest consumption of solvent-based adhesives is reached by installations for bus production with up to 100 tons per year. But the emission values might be exceeded together with the application of paints. For these sectors, techniques strongly reducing VOC emissions are already available.

3.1 Production of Passenger Cars

In this section, application of adhesives in the serial production of passenger cars will be discussed. Passenger cars are vehicles with less than 10 seats including the driver's seat. They serve for transportation of persons and luggage [Directive 70/156/EWG] (cf. also chapter 2.1.1, volume paint application). In 1999, 5.31 million of passenger cars were produced by the 7 German manufacturers [MÜLLER 2000].

Adhesives are processed punctiform in all production lines as opposed to varnishing. Within the last 10 years, solvent-based adhesives have almost completely been substituted (cf. chapter 3.1.2) [DECHEMA 1989]. As the situation in other EU countries might be different and the sector has processed huge amounts of solvent-based adhesives until a few years ago, the German state-of-the-art will be discussed in the following.

3.1.1 General Information

In order to increase the torsion resistance and the passive safety of the car body, combined spot welding bondings (instead of pure welding joints) are created. Application of modern adhesives and sealing agents in car body construction allow long anti-corrosion protection guaranties, as corrosion protection layers are not damaged by adhesives bonding and crevice corrosion is inhibited resulting from the sealing effect [VERBAND DER KLEBSTOFFINDUSTRIE 2000].

Adhesive bonding provides the possibility to join parts distortion-free. A post-treatment of the surface as it has to be carried out for example after spot-welding, is not necessary and a higher surface quality can be achieved. By applying adhesives as adhesive beads, greater production tolerances can be bridged. Elastomeric adhesives can be used effectively for embankment of vibrations during vehicle operation and in order to improve acoustic characteristics [ADHÄSION 1997]. A variety of adhesives is used as bonding material as well as sealing agent in the car construction sector. Often a precise classification (sealants / adhesives) of applications is not possible [HINTERWALDER 1991].

As a result of the EU Ordinance on old cars, producers try to construct the vehicles in a way that a trouble-free disassembly can be carried out after the usage period. Adhesive bondings require high efforts for separation hence they have been replaced by mechanical bonding techniques in some areas.

The application of solvent-based adhesives does not have any significance in the serial car production any more. Due to technological aspects (e.g. splitting off of acids during spot-welding adhesive bonding), health protection reasons and as a result of increased regulations about inside emissions of new vehicles, a reduction of solvent-based adhesives has taken place.
Further parameters causing a reduction of solvent-based adhesives are especially the following:

- Cancellation of adhesive techniques for wall-to-wall carpets due to the use of foam backed moulded parts.
- Substitution of adhesive bondings at insulating materials and facing parts by mechanical bonding techniques.
- Substitution of adhesive bonded rubber seals by mechanical fortifications.
- Substitution of adhesive systems by solvent-free adhesive systems for car body construction (body shell) due to fire protection reasons and due to avoidance of corrosive fission products of the adhesives while heating by spot-welding.
- Increasing shift of production processes towards enterprises of component suppliers.
- Substitution of adhesive bondings by mechanical fixations due to an improved possibility for removing parts after the usage period.

The hot-setting adhesives generally cure in the dryer installations of the varnishing department. Volatile fission products that may arise during this process are leaded to post-combustion installations together with the exhaust gas.

3.1.2 Applied Processes and Techniques

3.1.2.1 Application

Application of materials in the car production is mostly carried out line-shaped as strings or as beads for highly viscous materials. Application by the injection nozzle is realized with support of the material’s hydrostatic pressure created within the barrel press (“airless” method). Since the material is not sprayed by compressed air no overspray is generated. The application nozzles are formed according to the geometry of the adhesive bead. For plane application, the material is sprayed hot under low pressure. Dispersion adhesives are applied via compressed air or airless. Application devices are partly operated manually, however adhesive application is predominantly carried out automated by robot installations.

Hot melts typically exist as granulates or skeins, as cartridges or barrel fillings. Processing of the solid material depends on the form of delivery and the processing device. Before processing, the materials are melted. Application devices vary from hot melt guns with built-in melt accumulators, bigger units and barrel melting installations with heated ground plates. The melted material is supplied for the application by heated flexible tubes under defined temperature conditions [ENDLICH 1997].

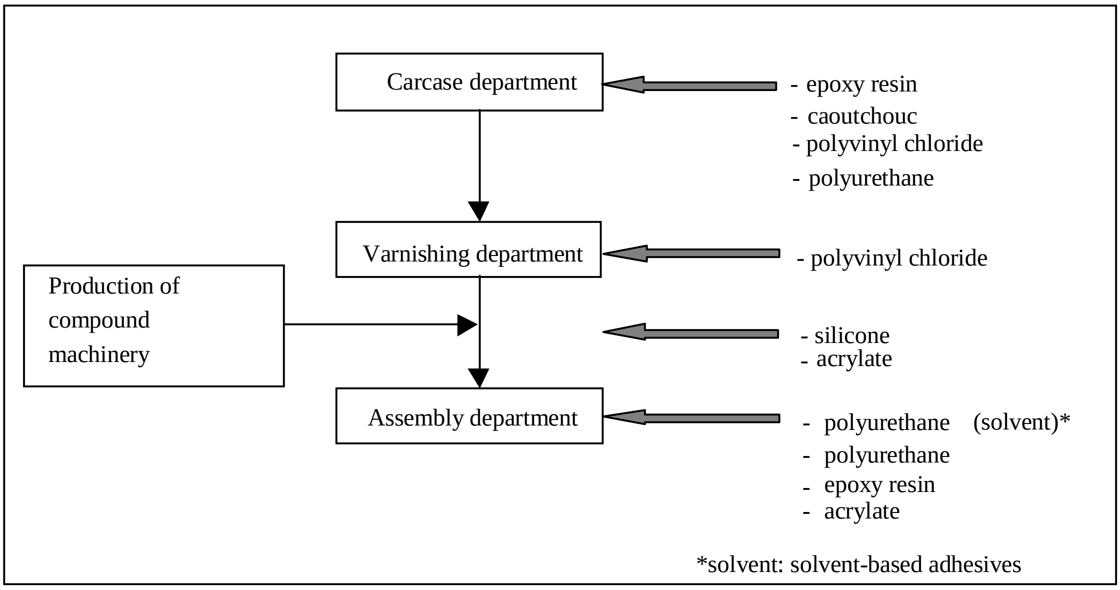
Paste-like adhesives such as the so-called shell construction plastisols are typically supplied in barrels. For the output of these highly viscous materials, hydraulic or electric barrel pumps are used. Partly, the pressure stamp and the feed pipes leading from the barrel press to the application areas are heated in order to improve the flowing capability of the highly viscous materials.

Certain mass ratios of individual components are supplied by barrel pumps for two part adhesives. They are mixed in a mixing installation (e.g. mixing pipe or dynamic mixer) and supplied to the application areas. One part adhesives cure by reactions with air humidity or by heating in the dryer installations (for example in the varnishing department).

3.1.2.2 Production Lines with Adhesive Application

Adhesive applications in the serial car production can be found in the production lines body in white construction, varnishing department, aggregate production and assembly (see fig 3-1). The motors and transmission units produced in the aggregate production section are obstructed in the assembly department. Fig 3-1 shows the production lines with adhesive application.

Figure 3-1*: Production lines with adhesive application in the serial production of passenger cars*



The applied techniques vary between different producers mostly with respect to the automation of level of adhesive application. However, good conformities can be found concerning the use of different adhesive types. In the following, techniques applied in the serial vehicle construction will be described.

3.1.2.3 Carcase Department

The components of the car body are welded and/ or cemented in the body in white department. For this process, solvent-free adhesives are used, especially epoxy resin adhesives, caoutchouc adhesives and polyvinyl chloride seam sealing agents. These materials are used as spot-welding adhesives, caulking bulks and as metal composite adhesives.

The adhesives processed for the mounting parts such as doors or shutters (or the whole car body for some producers) are pre-gelled in a dryer for 20 minutes with at temperature of 140 °C in order to achieve a sufficient washing resistance of the compounds in the production process of the varnishing department. For some mounting parts also induction installations are used. For the inductive pre-gelling, a current is induced in the folding area of the mounting parts that will heat the metal and the adhesive. The volatile components of the adhesives (plasticizers etc.) emittedg during the pre-gelling in the dryer installations are leaded to a post-combustion unit (generally of the varnishing department) together with the exhaust gas. Final curing of the adhesive compounds is carried out in the dryers of the varnishing department. The techniques used in carcass can be classified as **structural adhesive bondings** and **supporting and lining adhesive bondings.**

Structural Adhesive Bondings

Highly stressed parts of the car body such as door frame and door skin are bonded structurally (in other words strongly stress resistant). The structural bonding of the doors is realized without spot welding bondings. Adhesive application is carried out by eddy spraying technique (airless) along the fold seam profile. After bonding of door frame and outside panel, the door fold is beaded automatically by so-called beading presses. Also elements of the car body such as parts of the wheel case or at some producers the complete side plating are also bonded structurally. Adhesive application is carried out in the same way as for the door production.

Hot curing one part adhesives or epoxy adhesives are used as structural adhesives. Polyurethane adhesives are not in use any more as they emit toxic fission products when heated (e.g. welding). The advantage of epoxy adhesives compared to induction techniques is that their curing is completed within seconds so that a firm connection of parts can be guaranteed. Processing requires higher installation effort as mixing equipment is necessary.

Supporting and Lining Adhesive Bondings

Supporting and lining bondings are realised with polyvinyl chloride plastisols or caoutchouc adhesives. For example, roof and door hoops[1] are supportively cemented. Adhesive beads are applied in many sectors in between metal sheets laying one upon the other in order to avoid vibrations. Hood hoops are cemented with hot-setting caoutchouc adhesives by some manufacturers.

Tank connecting pieces and the engine bonnet hoop are fixed with solvent-free hot-setting caoutchouc adhesives. Application and installation are mostly carried out automatically.

3.1.2.4 Varnishing Department

After passing the cataphoretic dip coating installation, the seams and beadings of the car body are caulked due to corrosion prevention. By that, free punches, sheet doublings as well as spot welding seams are to be sealed against penetration of moisture as these can not be sealed by the ground coat. This prevents of possible corrosion. Polyvinyl chloride plastisols and reactive hot melts are used for caulking of the car body. Coarse caulking is applied automated by robots in the areas that will not be visible after the complete assembly process. The visible fine coating (such as door beadings, trunk area) is often applied manually by supplying the adhesive caulking via hose lines or out of cartridges.

[1] Bars for reinforcement of the car roof and the doors

The sound-absorbing mats are fixed either mechanically, by self-adhesive coatings or via a hot melt layer. These have been applied onto the materials already by the component supplier enterprises. These adhesives are re-activated by heat in the varnish dryers or the varnishing department and joined onto to the car body's surface that consists of the cataphoretic dip coat surface.

3.1.2.5 Assembly Department

In the assembly department, the vehicle is completed with drive assembly, interior decoration and windows. In comparison with other production lines, the biggest amount of adhesives is processed for glass metal connections in the assembly department. Two part polyurethane adhesive systems are applied whereas one or two part hot curing epoxy resins, hot curing butyl caoutchoucs reactive hot melts or methaacrylates are used for plastic bondings. These compounds are solvent-free.

Some manufacturers cement the roofing velt and the car sky with polyurethane adhesives. The applied adhesive bead serves as a support for the complete construction besides the bonding of car sky, roofing velt and roof. Due to the capability of parts for recycling intended nowadays, most fixations are carried out mechanically.

Fixation of the control panel on the splashboard is carried out partly by combined screw-adhesive bondings with moisture cross-linking polyurethane adhesives. Application of the Adhesive bead is automated. Partly, spare wheel troughs made of plastic are cemented into the car body in the assembly department with one part polyurethane adhesives. Processing and cementing are carried out automatically. The inner door foil is cemented with butyl caoutchouc (hot-butyl).

The immobile **windows/glasses** are cemented by moisture curing of one part moisture cross-linking polyurethane adhesives or reactive hot melts. For priming of glass, an application of a solvent containing primer (1 to 2 grams per vehicle) is necessary. The window frame of the car body is optionally cleaned manually with organic solvents (spraying bottles, rags). This removes the overspray originating from the cavity sealing.

Production and installation of so-called **cable trees** is carried out with the help of adhesive tapes. Cable transits through the splashboard towards the vehicle's interior are sealed adhesive-free by mechanical fixations.

3.1.2.6 Production of Aggregates

Surface seals of motor and transmission parts are realised by solvent-free, moisture curing silicone adhesive caulkings or duroplastic hot curing caoutchouc adhesives whereas screw joints are protected from loss of terminal strength and loosening due to vibrations by anaerobe curing methacrylates.

3.1.3 Current Consumption and Emission Values

Table 3-1 gives an exemplary overview over adhesive application in the production lines of the serial production of passenger cars, exemplary illustrated for compact car.

The adhesives used in carcase, varnishing department and assembly are one part adhesive caulkings curing by heating in the dryer installations, by induction installations (e.g. mounting parts) or by air humidity.

The total amount of adhesives processed per vehicle amounts 2.5 - 4 kg depending on vehicle size and type. Far between in the serial production of modern cars, about 10 - 20 g solvent-based adhesive caulkings per vehicle are processed for very few car types. Application of these adhesive systems is limited to small special purposes. For example, a flange within the power train is sealed against oil losses with a solvent-based polyurethane caulking or the battery case lit is laminated with carpet by one producer. No application of solvent-based adhesives is intended for the future car generation.

Table 3-1: *Application of adhesives in the serial production of passenger cars exemplarily for the AUDI AG* [KAULBERG 2001]

Application purpose	Material basis
Carcase department	
Adhesive for flanged seams- and spot-welding	Epoxy resin
Adhesives for fanged seams – and spot-welding for aluminium	
Structural adhesive	
Structural and beading adhesive bonding, resistant against washing out	Caoutchouc
Supporting and lining adhesive	Caoutchouc, acrylate, polyvinyl chloride / epoxy resin, polyurethane
Varnishing department	
Sealing of delicate seams	Polyvinyl chloride, solids content about 97 %
Sealing of coarse seams	Polyvinyl chloride, solids content about 97 %
Assembly department	
Adhesive for glasses	Polyurethane (air humidity curing)
Glass activating agent*	Silane, ethanol > 95 %
Glass primer*	Polyurethane, acetates und ketones about 60 %

*solvent-containing products

3.1.3.1 Adhesive Wastes in the Passenger Car Serial Production

Following wastes are created by operation and maintenance of the application installations:

- Overspray from adhesive application via spraying technique
- Adhesives remaining in containers
- Cleaning cloths with adhesive leftovers

- Outdated and waste adhesive mixtures

Cured epoxy adhesives can be disposed as industrial waste whereas polyurethane adhesives are hazardous waste.

3.1.4 Emission Abatement Measures

Predominantly solvent-free adhesives are processed in the passenger car serial production excluding a few small special applications. An application of solvent-based adhesives is not intended for the newer vehicle generations. A further reduction of VOC emissions in installations of adhesive application in the vehicle industry is not possible. The applied technologies can be classified as already established BAT. Due to increasing application of moisture curing adhesives, energy expendable drying techniques are reduced.

3.2 Production of Commercial Vehicles

3.2.1 General Information

Commercial vehicles serve for transport of passengers and goods. In 1999, 366.343 trucks and 11.825 buses were produced in Germany. Compared the serial production of passenger cars, the level of automation is significantly lower [Rippert 1998]. In the truck production, mechanical bonding techniques are predominant. Solvent-free adhesives are exclusively processed in the production of driver´s cabs for trucks.

Emissions arise in processing and drying of solvent containing adhesives in the bus production. Analogically to the passenger car serial production, low-emission adhesive techniques are used in the production of commercial vehicles. Predominantly moisture-curing adhesives are applied in the bus and truck production. Besides the avoidance of emissions, these adhesives are also energetically favourable as no dryers are necessary for curing of the adhesives. *An exception is cementing of the polyvinyl chloride floor cloth onto the wooden floor in city and travel buses and cementing of insulation of the engine compartment, edge protection and dense medium mats for some producers. About 30 - 35 kg solvent containing adhesives are used per vehicle.* In the production of truck cabins, no solvent containing adhesives are used at all in the German plants (see also chapter 3.2.2.3).

Due to the low quantities and the partly realised substitution of solvent-based adhesives, the sector of the production of commercial vehicles is not in the scope of the Directive. However, together with the processed varnishes and cleaning agents, the threshold values of the Directive are exceeded in some installations for bus production. The varnish processes and adhesive applications take place spatially separated.

3.2.2 Applied Processes and Techniques

Adhesives are used in the bus production lines carcase, varnishing department, assemble department and for mounting parts. In the bus production, side walls, roof skin, front and rear panelling, mounting parts made of sheet metals or plastic as well as floor cloth are cemented.
Adhesive application in the bus production is carried out manually out of barrel installations or out of cartridges. There is no automated application also due to greater production tolerances originating from the construction compared to passenger car production.

Similarly to the passenger car serial production, the interior decoration (seats, door linings, instrument boards) is acquired from component supplier enterprises and obstructed in the assembly line. Hence the adhesive technical processes do not take place at the car manufacturer any more (cf. 3.4).

3.2.2.1 Carcase

Carcase of buses

Bus production is distinguished from passenger car production by the significantly lower automation level. Buses are produced in smaller numbers.

Structural and supporting lining bondings are applied onto the (cataporetic dip coating) grounded bus carcase. Moisture-curing one part polyurethane adhesives and one part polyoxipropylenes, so-called MS polymers (**M**odified **S**ilicon) are used for structural adhesive bondings. MS polymers are used instead of polyurethane adhesives for cementing of zinc-coated metal sheets, as these show a broader adhesion. MS polymers are free of isocyanate and are not in the scope of the labelling duty. Side walls of the vehicle are fixed with MS polymers and lined with butyl caoutchoucs. Front and rear panelings, tail seats, inside panelings and the roof skin consisting of glass-fibre reinforced plastics (GFP) and are cemented with polyurethane adhesives. Adhesive application is carried out manually with barrel installations or cartridges.

Carcase of trucks

Structural cements and lining bondings in the carcase of truck cabins are carried out with one part caoutchouc adhesives. The side metal sheets of the cabin are bonded to the frame. Adhesive curing takes place in the drying ovens of the varnishing department. Adhesives are applied automated as a so-called adhesive bead. Extraction of the adhesive out of the barrel installations is carried out by hydrostatic pressure that is created by the barrel pumps following plate.

3.2.2.2 Varnishing Department

Buses/Trucks

Similar to the passenger cars serial production, caulking of welding seams and sheet doublings is carried out in the varnishing department with polyvinyl chloride plastisols or one-and two part polyurethane adhesives.

3.2.2.3 Prefabricated Construction

Prefabricated Construction of Buses

In processing of interior fittings, moisture curing one- and two part polyurethane adhesives as well as neoprene adhesives are used. One part polyurethane adhesives are utilised for cementing of vehicle windows and the plywood floor. Splices of the wooden floor plates are sealed with two part polyurethane adhesives. Some producers glue a polyvinyl chloride coating onto the plywood floor for city busses. This is carried out with solvent containing neoprene adhesives. Small amounts of solvent containing neoprene adhesives are used for cementing of insulation of the engine compartment, for edge protection and for dense medium mats by some producers (see table 3-3). About 30 - 35 kg of neoprene adhesives are processed per vehicle (solids content about 20 %) [JUREK 2000]. Some producers already process alternative adhesive systems [EVOBUS 2000].

Interior Fittings of the Truck Cabin

Interior fittings include cementing of the front module (instrument board), vitrification and the roof. Moisture curing one part polyurethane adhesives are processed for these adhesive bonding techniques. These products contain isocyanate. Therefore the exhaust gas is sucked off and is lead away by the roof. Application of adhesives is carried out automated. The adhesive plane of the glass parts is coated automatically with a solvent containing primer (about 5 g per vehicle). The cemented roof is joined with a graphite iron as stiffener. The total amount of adhesives amounts to about 2,5 kg per truck cabin.

Production of the Superstructure for Trucks

Solvent-free two part polyurethane adhesives are processed for cementing of the wall elements of the cooling superstructure. For assembly of wall elements, two part polyurethane adhesives or moisture curing one part polyurethane adhesives are used [FUHRMANN 2000, KLEBEN-DICHTEN 8/99].

If superstructures for vehicles build for transportation of dry goods are produced, solvent-free one or two part polyurethane adhesives are used for cementing of the single segments. Adhesive application is carried out manual.

3.2.3 Present Consumption and Emission Values

Since no solvent-containing adhesives are applied in the truck production, there is a description of the adhesive amounts processed in the **bus production** in the following.

3.2.3.1 Emissions in the Air

Some producers still use solvent containing neoprene adhesives for cementing of floor clothes in travel and city buses as well as for cementing of isolations and sealings. The mass of VOCs emitted from the adhesive application amounts up to 24 - 28 kg per produced vehicle. The emitted VOC mass may be reduced to 2 kg per vehicle if alternative techniques are used. With a maximum production of 10 vehicles per day, the threshold values for the consumption capacity of solvents originating from adhesive application specified in annex I (6.7) of the Directive have nor been reached by any one of the producers. However the threshold values are exceeded in some installations when together with the solvents originating from varnishes and cleaning agents are accounted for.

3.2.4 Emission Abatement Measures

In the following, process-integrated measures for VOC emission abatement will be presented. They are applied in serial production at the moment and therefore they can be classified as best available techniques.

Spray application of floor clothmaterial in city buses

As an alternative to the cemented polyvinyl chloride floor cloth, the floor material may be injected as solvent-free two part polyurethane material in order to avoid VOC emissions in the production of city buses [PREISSIG 2001]. This technique is favourable as the floor cloth has to be resistant against mechanical stress (resistance against footsteps, gliding and abrasion) and it has to prevent migration of moisture into the wooden floor besides the decorating effect. Besides the avoidance of solvent emissions, the use of polyvinyl chloride is waved. The material price is about two times high, however it may be compensated by an easier processing as well as by a reduced amount of claims [EVOBUS, SIENER 2000]. Due to the seam-free coating of the inside part of the vehicle, a closed winnow is formed that prevents the wooden floor better of penetration of moisture than cemented material (with seams). Repairs of single imperfections are possible. Cured leftovers of the decorating material can be disposed of as industrial waste. The non-slip properties of the material are better compared to polyvinyl chloride.

The application procedure includes the following working steps:

- The decorating layer is injected onto the sealed and polished car floor. The material is a solvent-free two component polyurethane material. Creation of colours depends on the customers' requests. The applied layer is about 2 mm thick. For a vehicle of 13 m length, about 85 kg are applied. The application is carried out manually with the air mix technique.
- For special effects, a tinseling material may be applied onto the decorating material if it's still wet. The application is carried out manually with low-pressure spray guns. The material is dried in a dryer at about 80° C for half an hour in order to reduce the drying time [PREISSIG 2001].

Recycling of the material after the usage period of the car has not been possible yet however it is being developed [EVOBUS, SIENER 2000].

Cementing of the floor cloth with adhesive dispersions with water as dispersing agent

Cementing of polyvinyl chloride floor cloths in the passenger area may be carried out in the plane with adhesive dispersions. Close to the wheelhouses and other strongly domed surfaces, a solvent containing neoprene adhesive is applied in order to counteract the restoring forces of the material. Presently one producer applies the technique [EVOBUS, SIENER 2000].

Substitution of insulating material mats by a two componenet polyurethane coating

The dense medium mats cemented with a solvent adhesive are replaced by a two part polyurethane insulating coating. Due to gap-free application at relevant spots (no joints), a reduction in weight could be achieved while the required acoustic insulation were maintained.

Table 3-2 shows currently applied techniques in order to save solvent adhesives exemplary for the EVOBus company. These techniques may be classified as established best available techniques. These techniques are confronted with conventional techniques in the table. The consumption of solvent adhesives may be reduced from 15 kg to 2 kg per bus for city bus production with these measures [EVOBUS, STEINER 2000].

Table 3-2: *Reduction of solvent adhesive application at the EVOBUS enterprise* [EVOBUS]

Application	Conventional processing	New techniques	Savings in adhesive [t/y]*
Installation of engine compartment isolation	Contact bonding with solvent-based adhesive	All isolating parts are equipped with a self-bonding acrylate adhesive	3.2
Installation of dense material pads	Contact bonding with solvent-based adhesive	Two part polyurethane dense material coating	4
Installation of floor covering, vertical surfaces, tail seats, wheel arches	Contact bonding with solvent-based adhesive	Two part polyurethane decorating coating	4.8
Installation of shutter caulkings	Cementation of tailored sealings (vulcanised edges) with solvent-based adhesive	Usage of an infinite profile equipped with a self-bonding acrylate adhesive	0.8
Installation of edge protection for wheel arches	Cementation with polyurethane in excess followed by cleaning with solvent	Cementation with a combination of polyurethane adhesive and an acrylate adhesive tape	0.9

*The savings in adhesive arerelated to 1600 bus units that were produced in 2000

3.2.5 New Progressive Techniques

Solvent-free adhesive systems for a complete cementing of the floor cloth are currently in the development stage [PREISSIG 2001].

3.3 Production of Trailers and Mobile Homes

3.3.1 General Information

In 1999, 60.243 trailers and 21.910 mobile homes were produced in Germany from 53 companies.

Solvent-containing adhesives are only used by a few producers for lamination of small areas in the trailer and mobile home production. Therefore it is not relevant with respect to the Directive. All bigger adhesive bonds are carried out with solvent-free adhesives [PRIEBE 2001, KLEBEN/DICHTEN 12/1999].

3.3.2 Applied Processes and Techniques

Moisture curing polyurethane adhesives are applied for cementing of side walls and the roof of trailers and mobile homes as well as for fixing of water tank fasteners [PRIEBE 2001]. The adhesive systems do not need any additional energy supply for curing. Adhesive application is carried out manually out of cartridges or barrel installations. The sandwich elements of the wall construction and of the roof are cemented by solvent-free, moisture curing one part polyurethane foams. The sandwich elements consist of aluminium sheets, polyurethane foam and plywood [PRIEBE 2001]. Seams and hems are sealed with polyvinyl chloride plastisols or they are laminated with caulkings.

3.3.3 Current Consumption and Emission Values

Solvent containing adhesives are only used by very few producers for small special applications such as lamination of strongly curved surfaces with carpeted floor (e.g. seat frame). With the exception of these applications, the branch does already use best available techniques. Therefore this sector will not be studied further.

3.4 Component Supplier Industry

3.4.1 General Information

A strong transfer of production processes towards the enterprises of component suppliers has taken place in the serial production of vehicles. The main application for adhesives are laminations of plastic facings with cloth for the interior decoration of the vehicle. Installations with consumption capacities higher than the threshold values specified in the annex I (6.7) of the Directive are presently unknown. Due to a situation of active competition originating from concentration processes, the sector was only studied exemplary at the Johnson Controls company. Other companies were not willing support any co-operation.

Emissions result from the application of solvent containing adhesives. Analogically to the passenger car serial production, a substitution of solvent adhesives has taken place in the component suppliers industry. Presently, heat stressed parts such as clutch and brake linings[2] are joined with solvent adhesives predominantly. Newer installations process solvent-free adhesive dispersions also due to the requirements of extremely low inside emissions of new vehicles. Solvent containing adhesives are used presently only in small amounts for production of leather linings [PRÖMPER 2001].

3.4.2 Applied Processes and Techniques

Reactive hot melts and watery adhesive dispersions are processed except by some older installations. The formerly used solvent-based neoprene adhesives have been substituted by one or two part adhesive dispersions in the adhesive bonding of cushion material and laminations [ENDLICH 1997]. Laminations of **indoor facings** are carried out with two part adhesive dispersions based on polyurethane or with hot melts based on polyamide and ethylene vinyl acetates as well as with reactive hot melts. The adhesive mass amounts to 60 - 80 g/m², depending on the type of adhesive and the combined materials.

Cementing of the material for the **trunk indoor facings** is carried out with hot melts. In the production of **door inside facings**, solvent adhesives have been substituted especially by watery adhesive dispersions based on polyurethane. The substitution has taken place due to stricter requirements of the passenger car producers concerning lower inside emissions of new automotives as well as for environmental protection reasons [PRÖMPER 2001]. The adhesives can be activated by heat and the parts being joined have to be cemented within a few hours after adhesive application.

Predominantly, adhesives are applied automatically by robots via compressed air spraying. The created overspray is absorbed dry in filter mats due to financial reasons.

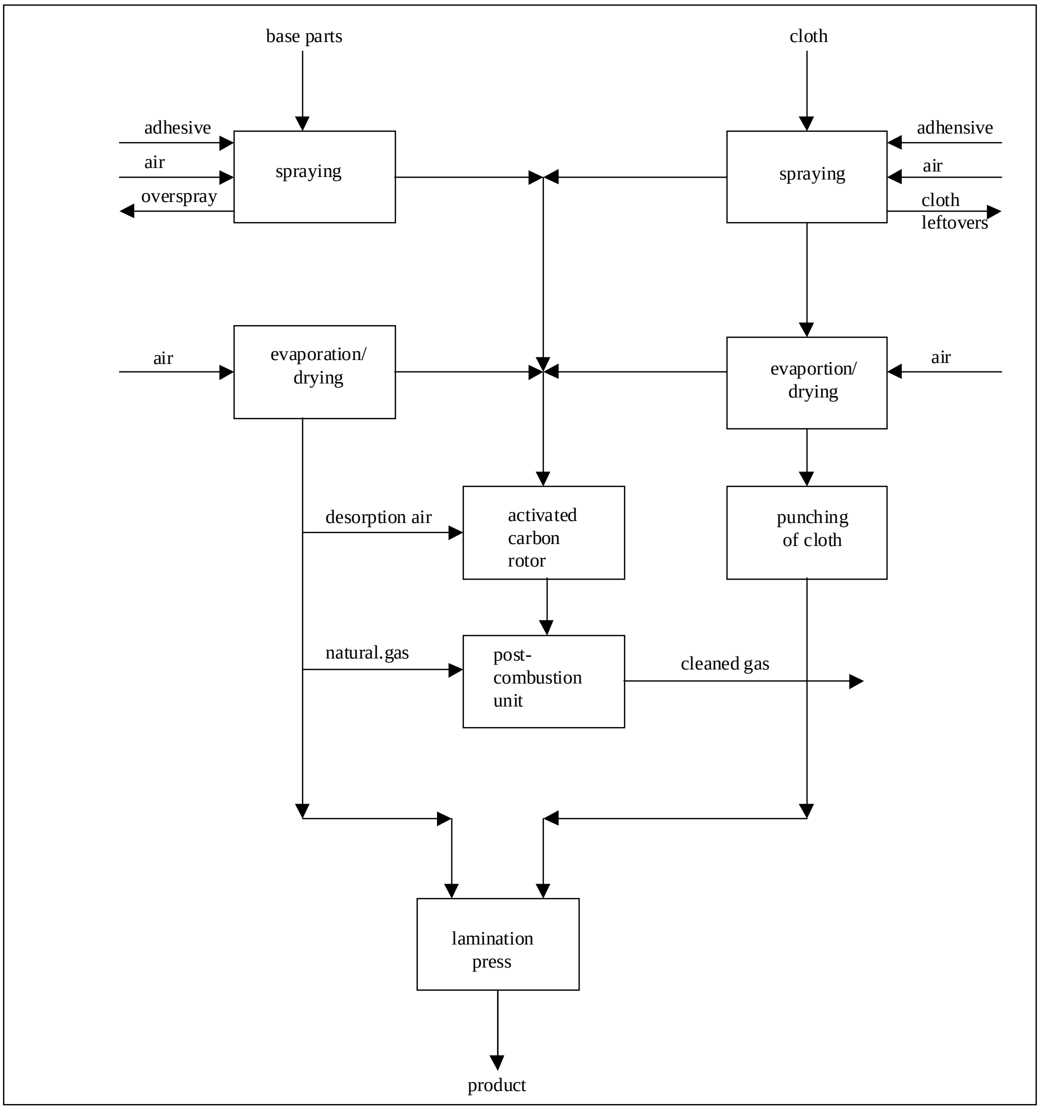


Figure 3-2: *Production of facing parts for passenger cars in conventional installations (center pillar)*

Fig 3-2 shows the production of facing parts for passenger cars by a conventional technique. The plastic parts being laminated with cloth and the cloth sheetings are coated with solvent containing polyurethane adhesive with about 20% solids content. They are applied via automated compressed air spraying.

[2] The production of clutch and brake linings is discussed in volume "Integrated protection of the environment in certain industrial activities - installations for impregnation, printing, saturating, coating"

Afterwards cloth sheetings and plastic parts are dried in a heated area. Lamination takes place in a lamination press. An activated carbon adsorption wheel captures the VOCs of the dryer and spraying booth exhaust air. The desorption air enriched in VOCs is cleaned by a post-combustion installation.

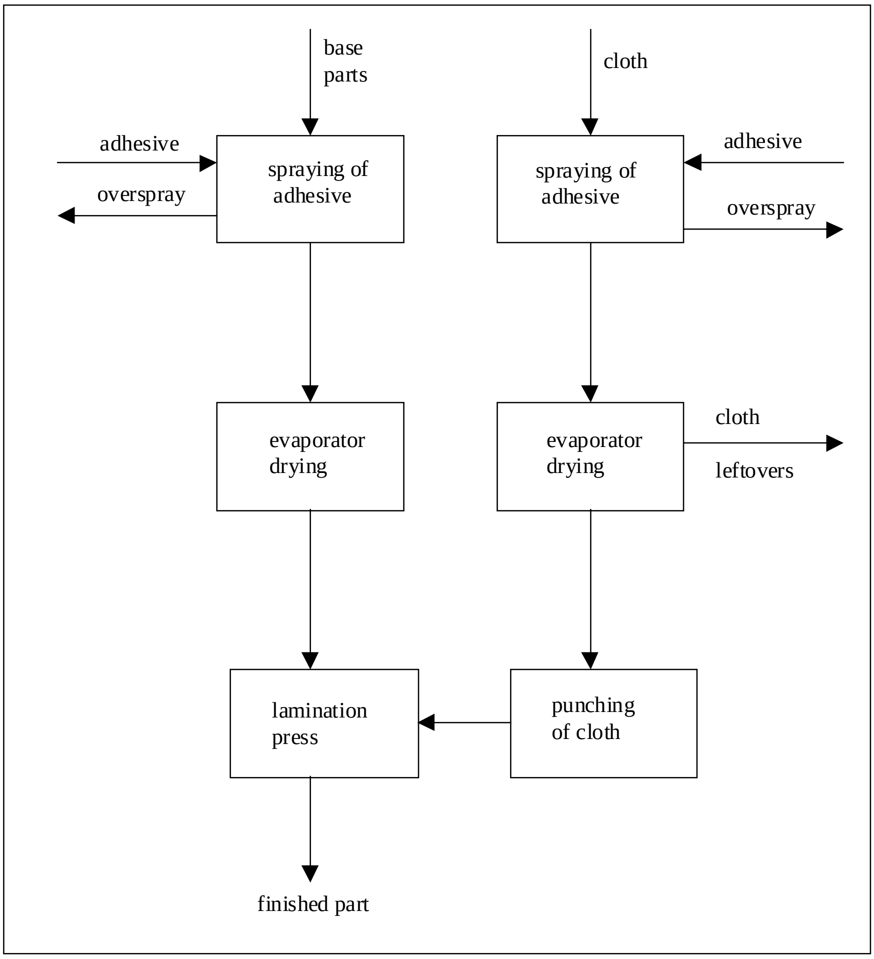


Figure 3-3: *Production of facing parts with watery adhesive dispersions*

Fig 3-3 shows the lamination of door facings exemplarily for the Johnson Controls company. A watery adhesive dispersion based on polyurethane with isocyanates as curing agent is used as adhesive. All common inside parts with the exception of leather equipment may be laminated with this technique and adhesive system [PRÖMPER 2001]. Adhesive application is carried out with compressed air syringes. The overspray is separated dry.

For the production of leather seats and the lamination of instrument boards or door facings with leather in small serial production, solvent-containing polyurethane adhesives are processed (Solids content: About 60 to 80 %).

3.4.3 Emission Abatement Measures

Watery adhesive dispersions based on polyurethane are used for **lamination of indoor facings.** This adhesive system can be used for all common indoor facings with the exception of leather laminations according to the technique sketched in Fig 3-2. It can be classified as established best available technique.

For **leather laminations**, watery two part adhesive dispersions based on polyurethane can be applied in a (cold) contact technique for plane surfaces. Within the edge area, (so-called folded edges) and for strongly domed surfaces, solvent-based adhesives are processed exlusively. This is necessary to counteract the high restoring forces of the material.

3.5 Production of Rail Vehicles

3.5.1 General Information

New rail vehicles are made of metals and non-metals in a mixed type of construction (hybrid type of construction) in order to reduce production times and decrease weight. Therefore construction parts consisting of different materials have to be combined or bonded. Solvent-containing adhesives are not in use any more.

3.5.2 Applied Processes and Techniques

Applications for adhesives exist in all parts of the vehicle. They include adhesive bondings of facing parts in the underbody area (hinges, joints), in the complete front section (front window, front facings, front plate) as well as in roof mountings. The applications partly include bondings of supporting vehicle frames and the assembly of completely prefabricated side-wall systems. Adhesive bondings of frame structures, corner joints and side plates are carried out in container and wagon production as well as in the planking of containers and all types of caulkings as well as rubber seals for the special transport containers. No solvent containing adhesives are used for constructional (structural) adhesive bondings, but solvent-free one and two part polyurethane adhesives (PUR), hot melts (PUR), two part silicone adhesives and acrylate adhesives are used.

Nowadays, only rubber is used as floor cloth. The formerly used polyvinyl chloride is not applied any more due to fire protection reasons. For cementation of the floor cloth, solvent-free polyurethane adhesive dispersions are applied exclusively. These adhesives are processed with the contact bonding technique and allow a trouble-free removing of the floor cloth for repair works [ZIEMS 2001].

The technique can be classified as an already established best available technique.

3.6 Aircraft Construction

3.6.1 General Information

Subject of this chapter is the application of adhesives in the aircraft construction of civil air carriers with a minimum of 50 seats. Adhesive processes have a very low significance in the maintenance of aircrafts. Therefore in the following, only new construction of air carriers will be considered. The amounts of solvents processed in the adhesive processes and preparation steps are very low so that the threshold values of the Directive are not reached.

3.6.2 Applied Processes and Techniques

Adhesive application in the air carrier production has been investigated at the EADS Airbus enterprise at the Hamburg location. The installation produces about 70 aircrafts per year.

Structural adhesive bondings are in a strong competition with the bonding techniques welding and riveting in the production of commercial aircrafts. Fuselage sections and components of the wings are cemented. For newer constructions, mechanical bonding techniques are used increasingly [FANGMEIER 2000].

Nowadays solvent-free adhesives or adhesives with a low solvent content (solvent proportion < 2%) are processed as hot setting adhesive films or as paste-like two part epoxy adhesives in the structural aircraft construction [KARL 2000, KLEBEN/DICHTEN 12/1999].

Preparations of the surfaces being cemented are necessary for adhesive processes of the fuselage. The aluminium planking is prepared with chromic acid and is coated with a solvent and chromate containing primer (10 to 20 % solids content) in order to promote adhesion. The surface area being coated amounts to about 400 m^2 per aircraft. About 150 g/ m^2 are applied. At the Airbus Deutschland GmbH production site in Nordenhamm, about 70 tons of primers are processed per year and about 3 tons per year at the production site in Hamburg. [SCHRÖDER 2001, FANGMEIER 2002].

Presently no substitution is possible due to technological reasons such as compatibility with the adhesive system, long-term stability and corrosion resistance. Alternative water based primers are presently in a testing stage [SCHRÖDER 2001].

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4 Adhesive Application in the Footwear and Leather Goods Industry

4.1 Shoe industry

The German installations for shoe production doe not exceed the threshold values of the IPPC-Directive due to the sector's structure and due to the small amounts of processed solvent-based adhesives. However it has to be considered that installations of the shoe industry in other EU member countries with higher production amounts might reach the threshold values. Since there are no adequate statements about the European state-of-the-art of the shoe industry in the literature, the state-of-the-art of the German shoe industry will be described in this paper.

Subject of this chapter is the industrial production of shoes with a leather (shoe-) upper, where adhesive bonding is the most important joining technique [INDUSTRIEVERBAND KLEBSTOFFE 2000].

Due to the variety of applied adhesives, materials and bonding technique, the high requirements for the adhesive bondings as well as the relative comparability of the installations, the shoe industry is especially suitable for illustration of applied techniques in adhesive application.[1]

4.1.1 General Information

A street shoe consists of 50-150 parts. Soles, heel seats, toe puffs and heel stiffeners are manufactured separated by component suppliers [NECKERMANN 1987]. The parts belong to different material categories as for example cloth, plastics and leather. These materials differ in their technological features (such as surface properties) according to different origin and chemical pre-treatment.

Among the **synthetic materials** that are going to be cemented, polyurethane, elastomers and polyvinyl chloride have the biggest share [ADHÄSION 6/1999]. Besides these materials polystyrol, polyester, rubber and polyamide are processed.

In 1999, 39.84 million shoes were produced, thereof 31.48 million had a leather upper (80% of the total production) [HDS 1999].

Walking shoes (for women and men) have the biggest share with 51% (see fig 4-1).

[1] Basically it has to be mentioned that the installations applying adhesives vary strongly due to the sector´s heterogeneitys.

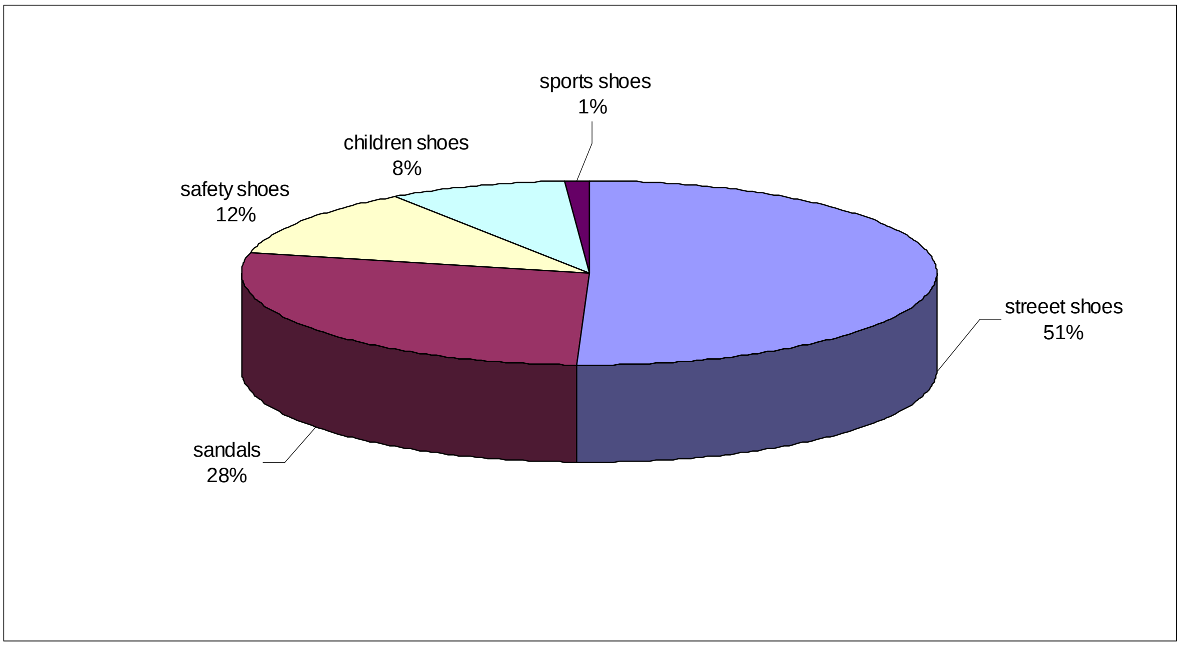

Figure 4-1: *Proportions of shoes produced in Germany in 1999* [HDS 2000]

The shoe production consists of small and middle-sized enterprises. Reasons for that can be found in the strong segmentation of the market by the enterprises and in the production of small batch sizes. Some exceptions are big producers (especially of sport shoes), characterized by serving a bigger market or having coupled production and marketing. [NECKERMANN 1987]. Partly more than 5000 pairs are produced daily in big installations.

At the moment, the German shoe industry is facing a **strong reduction of enterprises of all sizes.** In 1999, there were 147 shoe-producing enterprises in Germany with 17.779 employees. Since 1995 the number of employees was reduced by 19 % [SBW 1999]. The reason is the transfer of production capacities to Portugal, South East Asia and especially to Eastern Europe in order to reduce wage costs [ADHÄSION 4/1999]. Hence the share of imported shoes rises [GÜNTHER 1983, NECKERMANN 1987].

4.1.2 Applied Processes and Techniques

Manual labour is predominant in shoe production. Automation is only possible in some partitions of the production process (e.g. via the moulding injection) due to the big variety of forms and materials as a reaction on yearly assortment changes.

4.1.2.1 Adhesive Application

Solvent-based adhesives have a proportion of 67 % of the total consumption in the German footwear industry, adhesive dispersions and hot melts have proportions of 16 and 17%, respectively [ADHÄSION 6/1999]. The reason for the high proportion of solvent-based adhesives is the fact that the processed adhesives have to have a high bonding strength right from the beginning, since often materials under tension are bonded. The hardened adhesive bonds have to resist permanent mechanical stress and must not be sensitive to wetness. Often only solvent-based adhesives meet these requirements. Table 4-1 shows the adhesives processed in the footwear industry.

Table 4-1: *Adhesives in the shoe industry*

Dispersion in water	**Hot melt**	**Dissolution in organic solvents**
Polyurethane, natural and synthetic rubber	Polyester, polyamide, ethylen vinyl acetate (EVA)	Polyurethane, neoprene

Solvent-based adhesives are primarily used in the sole-top splicing. Polyurethane and neoprene adhesives (neoprene adhesives) are processed. The solid contents of the solvent-based neoprenes and polyurethanes vary in the range of 20 to 26 %. Especially ketones and esters (such as ethyl acetate) are used as solvents. The high proportions of organic solvents in the compounds are necessary for penetration of the adhesive into the leather fibres.

Hot melts are based on polyester-, polyamide- and vinyl acetate. The application depends on the processing machines and temperature sensitiveness of the materials being processed.

Adhesive dispersions are mainly based on polyurethane, natural or synthetic caoutchouc with water as dispersing agent. These adhesives are also referred as latex adhesives in the footwear industry.

Depending on the manufacturer and the shoe model, the total mass of adhesives used for the production amounts to 10 - 30 grams per pair.

Generally, **hot melts** are applied automatically by the stitching, lasting and beading machines. The adhesive is processed via jets onto the materials being joined. The application machines are supplied with the hot melt as granulate or skein. Immersion of toe and back puffs has the highest adhesive efficiency.

Generally, **adhesive dispersions** are applied manually by spraying techniques.

Solvent-based adhesives are processed either manually by paint-brushes or automatically by spreading machines. For the application of larger amounts of solvent-based adhesives, the workplaces and the spreading machines are equipped with exhausters.

4.1.2.2 Production Processes

Although there is a huge variety of the adhesive bonding processes, depending on the adhesive manufacturer and the level of automation, there are large conformities in the production flow as well as in the used adhesives. The use of a specific adhesive depends on the shoe type of construction and its materials. Fig. 4-2 outlines the process flow for production of Ago-type shoes. In the following, production processes with processing of adhesives will be described.

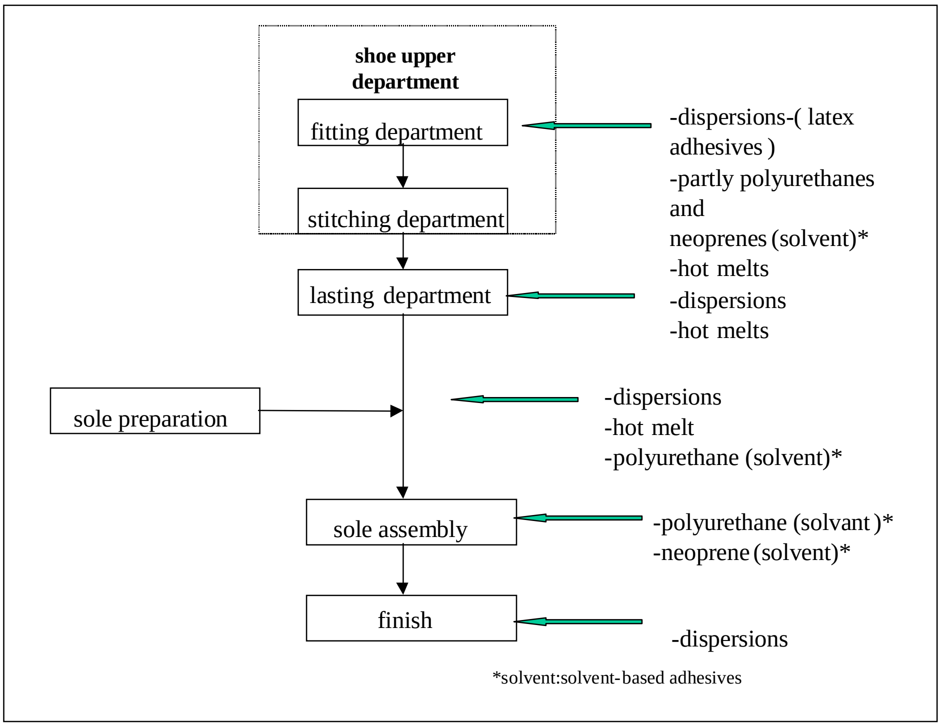

Figure 4-2: *Processing steps in the shoe production*

4.1.2.2.1 Shoe Upper Department

The shoe upper department comprises the fitting and the stitching department.
In the show upper department, components of the shoe upper as well as linings are connected by cementing and sewing techniques (stitching) to form the shoe upper. In the fitting department, components are fixed by adhesives before stitching. Application of adhesives is usually carried out manually via a paint-brush. Adhesive dispersions are applied wet via spray guns. Latex adhesives are used for cementing of shoe upper cushions and collar cushions as well as for bonding of vamp and quarter lining into the shoe. They are also applied for laminating processes of linings. Some manufacturers fix the collar and upper cushions with solvent free self-adhesive coatings. Besides that, reinforcing materials are bonded by heat compression. Reinforcing materials are textiles that have already been coated with hot melts (based on polyester, polyurethane, ethylene vinyl acetate or polyamide) in the component supplier's production. As reinforcement for single performed sews within the shoe upper, an additional bonding of the parts is carried out with solvent-based polyurethane cement. Generally, small seam allowances (overlap areas) can only be bonded with solvent-based polyurethane adhesives. The use of solvent-free adhesive dispersions is only possible for this application if the joining parts can be bonded over bigger seam allowances due to the lower adhesiveness and the lower tensile strength of adhesive dispersions.
Ethylene vinyl acetate and polyamide hot melts that are used for beading[2] are applied automatically by beading machines. Some manufacturers use hot melt layers as stiffener instead of conventional toe puffs. The adhesive is provided as granulate or skein according to the specifications of the machine.

Adhesive dispersions (latex) have a share of approximately 70 %, hot melts have a share of 20 % and solvent-based adhesives have a share of 10 % of the total consumption of adhesives processed in the shoe upper department.

4.1.2.2.2 Sole-fitting Department

Heel seats, orthopaedic socks, soles and insoles are concealed with leather or textiles in the sole-fitting department for later fitting in the sole assembly department. Cementing is carried out with adhesive dispersions, hot melts or with solvent-based polyurethane adhesives if the surface is strongly curved. For this process, the contact bonding technique is used (application on both parts being joined together, evaporation of the solvent cementation of the parts). Adhesives are applied manually by paintbrush or automatically by spreading machines.
For example shoe soles made of rubber generally need to be pre-treated with a halogenizer to ensure a strong adhesion of the adhesive layer onto the sole material. The application of these

[2] Folding back and fixing of leather edges in the shoe upper area

chemicals and adhesives is carried out manually under a fume hood. Afterwards, the soles are pre-treated with a neoprene adhesive as first coating that is dried after application.

The sole-fitting department processes solvent-based polyurethane adhesives almost exclusively.

4.1.2.2.3 Lasting Department

The shoe upper finished in the shoe upper department is attached to the insole and partly to the sole in the lasting department. Depending on the type of shoe construction , production steps differ. With respect to the use of adhesives, shoes have to be classified into different construction types: traditional **Ago type shoes**, **Strobel type shoes** and **Vertical Welt shoes**. Independent of the shoe construction, hot melt coated textile reinforcement materials (e.g. counters) are cemented by hot compression during the assembly process. Partly, counters are plunged into latex adhesives by the shoe manufacturer and the dried adhesive coating is re-activated by heat before assembling.

Hot melts and adhesive dispersions have a proportion of 50% of the total adhesive consumption in the lasting department, respectively. Solvent containing adhesives are processed for waterproof Ago type shoes only.

Ago Type

The insole and the shoe upper are mounted by so-called lasting on shoes of the ago-type. In this process, the lower edge of the shoe upper (lasting margin) is folded towards the inside and thereafter attached at the insole. For lasting the shoe upper is pulled tight and wrinkle-free over the last (a tool in shape of a foot to sculpture the shoe upper) until it has the form of the last. Thereafter, the lasting machine cements the insole with the shoe upper via the lasting margin. The hot melts are applied in the lasting machine (polyester and polyurethane). At the toe area of the shoe, polyester hot melt is used, but for the side parts of the toe the insole is attached with polyamide hot melts. Polyamide-hot melts are featured by a higher flexibility. The heel area can be attached (lasted) to the insole by different closing techniques. Children's shoes are lasted according to the described method. In some men's and women's shoes the fusion in the heel zone can be realized adhesive-free with tacks. If the fusion is not realized with tacks, polyurethane adhesives are used to cement the areas between toe area and heel.
Finally the sole is cemented with the insole or the insole is moulded with polyurethane, polyvinyl chloride or TR-plastics.

For ago type shoes that are sealed with a respiration active membrane, a special waterproof insole is built in. In these shoes the interspace between the insole and sole is sealed with solvent-based polyurethane adhesive. This extra sealing prevents water from penetration into the shoe.

Vertical Welt Shoes

At vertical welt shoes the lasting edge is attached to the insole over seams. The joint is realized without any use of adhesives. This type of construction is used for moccasins and hiking boots. The sole is attached to the insole seamed with the shoe upper. The sole is attached in the soil assembly department similar to Ago type shoes.

Strobel Type

At shoes of this kind the shoe upper is fixed to the textile insole with a special seam, the so-called Strobel-seam. The joint is realized without any use of adhesives. Fixing of sole and textile insole is carried out in the same way as in the Ago type. The Strobel type is used for sport- or trekking shoes for instance.

4.1.2.2.4 Sole Assembly Department

To attach the sole to the insole, adhesive processes and injection moulding are common techniques.

If an adhesive is used for joining sole and insole, solvent-based polyurethane adhesives are used and applied by a hot-setting technique. Solvent-based polyurethane (2 component) and neoprene (2 component) adhesives are used. Polyurethane adhesives are predominant in the total consumption of the sole assembly department with a share of 95%. Furthermore, neoprene adhesives are used for special applications, as for example for pasting of polyester splines into soles.

For the connection of sole and shoe upper by the **adhesive technique**, the shoe upper is pre-treated (precoat) with a polyurethane adhesive (2 component, 10% solid content). This precoat layer soaks in the leather surface and serves to harden the leather fibres. After evaporation of the solvents, a second layer of polyurethane adhesive (2 component) is applied (main coat). The joint of shoe upper and sole is carried out with a hot activating method. Sole and shoe upper are heated for re-activation of the dried adhesive and compressed hot. This re-activates the desiccated adhesive. The first cement layer is directly applied by the sole manufacturer (in case of rubber soles and ethyl vinyl acetate soles) and consists of neoprene and polyurethane adhesives.

The solvent-based polyurethane adhesives used for the sole cementing are featured by a larger grease resistance, a larger hydrolysis resistance and better adhesive strength if compared to adhesive dispersions and solvent-based neoprene adhesives. Another advantage of polyurethane adhesives is an unlimited time to be used. For sole assembly with the adhesive technique, solvent-based polyurethane and neoprene adhesives are applied.

With the **injection moulding technique**, the sole is directly moulded adhesive free onto the different models (Strobel type and Ago type). Materials for injection moulding used by the shoe manufacturers are polyurethane, polyvinyl chloride and plastics. The re-lasted shoe upper is enclosed by a sole-shaped mould filled with the liquefied sole material under high pressure. The liquefied sole material forms a strong bond with the shoe upper while it cools off. The processing operates without any organic solvents. Only for cleaning of the sole moulds of the rejection moulding machine, organic solvents are used.

4.1.2.2.5 Finish Department

In the finish department, orthopaedic socks are cemented into the shoe. Mainly adhesive dispersions (latex adhesives) are processed. For sandals the sole is joined with solvent containing polyurethane adhesives

4.1.3 Current Consumption and Emission Values

According to the Directive 1999/13/EG about the limitation of emissions of volatile organic compounds, the emission limit for total emissions is 25 g VOC per completed pair of shoes.

For the production of a pair mid-sized (42 or 8) walking shoes an amount of 20 to 60 g adhesives is used depending on the manufacturer. Solvent-based polyurethane and neoprene adhesives have an average proportion of 67% of the processed adhesives. Hence about 13,5 - 40 g solvent-based adhesives with a solvent's proportion of 75 to 80 % are used per pair of shoes. The emitted VOCs amount between 10 and 35 g per pair. The solvent use in big installations can be estimated about 11 - 39 tons per year. Due to the structure of the sector in Germany, the threshold values specified by the Directive are not reached by the existing installations in Germany.

Generally, a treatment of exhaust gases is not carried out due to the handcraft production processes. The application of adhesives in the shoe production does **not generate relevant quantities of wastewater,** because over spray is generally absorbed dry if a spraying application is used.

4.1.3.1 Wastes

Sources of adhesive waste in the shoe production are:

- Adhesive compounds descending from machine-cleaning processes.
- Spoilt adhesives, adhesive materials of which the assembly time was exceeded:

Approximately 10 % of the purchased solvent-based adhesives are disposed of as wastes.

- Over spray from spraying and other drop outs from paintbrush applications
- Adhesive barrels: Non-hardened contaminations of barrels have to be disposed of as hazardous waste. Hardened adhesive leftovers have to be disposed of as industrial waste. Partly, suppliers take barrels back. They are either used for recycling or as tinplate scrap.

The type of adhesive determines the amount of adhesives remaining in the empty container. Hot melt granulates can be used almost completely. In a 200 l barrel filled with highly viscous adhesives approximately 5 % will remain [ENDRIß, ELSLAND 1997]. The amount of waste can effectively be reduced by installations for emptying of residues.

A higher automation level of the adhesive coating process does not necessarily reduce the amount of waste. Highly automated processes can generate a large quantity of wastes as well. Spray coating with adhesives produces more wastes than other application techniques due to over spray [ENDRIß, ELSLAND 1997].

4.1.4 Emission Abatement Measures

In the following, potential measures for primary emission abatement are described:

- Use of solvent free adhesives

- Use of adhesive free soling techniques

- Use of exhaust cleaning installations such as post-combustion, bio-filtration or adsorption.

- Consumption effective adhesive systems, use of devices that allow a total emptying of the adhesive containers or use of inliners.

4.1.4.1 Use of Solvent-Free Adhesive Systems

Most of the solvent-based adhesives are used for bonding of sole and shoe upper. Alternatively, sole and insole can be cemented with solvent free adhesive dispersions based on polyurethane or via hot melts.

Water based adhesive dispersions are mainly based on polyurethane. These adhesives are more difficult to handle than conventional solvent-based adhesive systems. A disadvantage in comparison to solvent-based adhesives is the short assembly or open time. A long-term storage of adhesive dispersion coated parts is not possible. The parts need to be assembled directly after the adhesive application. A reactivation by heat as with polyurethane and neoprene adhesives is not possible. A save bonding with adhesive dispersions is only possible for certain material combinations. Up to now, the diversity of material combinations used in the production of fashionable shoes does not allow the use of adhesive dispersions [ZIEGLER 2000]. Problems in the application of adhesive dispersions are their low starting strength and their poor surface penetration. However a high starting strength is necessary for bonding of soles and other curved materials to bear down the restoring force [ADHÄSION 6/1999].

Manual and automatic paintbrush applications of water based adhesive dispersions is problematic, because the adhesives stick to the paint-brush and it will desiccate quick. Therefore the paintbrush has to be replaced often and this increases the work effort [WOLTER 2000]. Adhesive dispersions can be applied best with spraying techniques. Spraying of adhesives can be of advantage compared to manual paintbrush application because it may require less time. However the use of water based adhesive dispersions as thick coatings is associated with longer drying times even if drying installations such as ovens or tunnels are installed. This is due to the higher evaporation enthalpy compared to organic solvents and the slower penetration of shoe material by water [ADHÄSION 6/1999].

The cycle time is about 30 - 40 minutes longer compared to solvent-based adhesives even with the use of drying ovens depending on the adhesive and materials being cemented [URBAN 2000]. To avoid disadvantages, water based adhesive dispersions have to be applied very precisely. In spite of the described disadvantages, adhesive dispersions based on polyurethane or neoprene are used as main adhesives besides classical applications in the serial production by some manufacturers (e.g. Nike). Besides ecological and sanitary advantages, adhesive dispersions are featured by a higher bonding strength after drying compared to solvent-based adhesives [ADHÄSION 6/99]. Furthermore, the use of adhesive dispersions also reduces the required storage capacities due to the solid content of the products that is 3 times higher.

For the splicing of soles, reactive hot melts can be processed [ADHÄSION 6/99]. These adhesive systems require an exact compliance of process parameters such as temperature, moisture content and pH. Therefore, processing of these hot melts is problematic

[ADHÄSION 6/99]. Hot melts are used as a solvent-free alternative in the production of gym shoes [URBAN 2000]. These hot melts can be used for bonding of shoe upper and sole because plane (not curved) soles are used in the gym shoe sector. For this sole type, the application of hot melts by rolls is economic.

4.1.4.2 Adhesive Free Sole Assembly

Injection Moulding

Adhesive free sole assembly with injection moulding of fluid polyvinyl chloride or polyurethane can only be implemented for large batch production due to the high investment needs (closed automated installations and moulds) and considerable operation costs. Another disadvantage of injection moulding is the limited variety of sole shapes and colours. Therefore injection moulding is mainly used by manufacturers of sports shoes and children shoes [ADHÄSION 4/1999].

Sewing techniques

In sewing techniques, sole and insole are sewed up manually. The attachment of the sole with this technique is carried out without any use of adhesives .

4.1.4.3 Use of Exhaust Gas Treatment Installations

Due to the low concentration of VOC in the exhaust gas, thermal exhaust treatment installations are not applicable [HDS 2000]. For large batch production, bio-filtration processes might be applied [ESIG 2000]. With respect to the operating costs, bio-filtration is less expensive than post-combustion due to low VOC concentrations in the exhaust gas [ESIG 2000]. Presently, exhaust cleaning installations are not in use in Germany.

4.1.5 Selection of Emission Abatement Measures

- Substitution of solvent-based adhesives by hot melts or adhesive dispersions for sole and shoe upper bonding. Processing parameters have to be fulfilled precisely which results in a higher processing effort. Due to the variety of processed materials of fashionable shoes, the application of adhesive dispersions has not been possible in Germany up to now. Longer cycle times as well as investments for additional drying installations are necessary for the application of water based adhesive dispersions.

- Injection moulding of soles instead of Ago or Vertical Welt type shoes. The technique is limited in its application in the fashionable shoe sector due to limited sole forms and colours.

- Capture and bio-filtration of solvent containing exhaust gases if solvent concentration in the exhaust is high enough (also for reduction of odour emissions).

4.1.6 New Progressive Techniques

Some adhesive manufacturers are currently developing solvent-based adhesive systems with a solid content of approximately 60 % for adhesive processes of shoe upper and sole. Also, water based first coatings of cement for soles are being developed. These products have not been used in the production yet. [FOBRO HELMITIN 2000].

4.2 Leather Goods Industry

In this chapter the production of leather goods is described. Products of the leather industry are bags, products for travelling, purses and other small leather goods. Leather goods are fashionable products like shoes made out of various materials. Different sewing and adhesive processes are applied in the production.

In 1999, 300 enterprises with 5960 employees worked in the production of leather goods in Germany. Thereof about 100 enterprises had more than 20 employees. The sector mainly consists of handcraft enterprises. In 1995, only 150 enterprises working in the leather goods production remained. The reduction of enterprises was partly caused by migration outside of Germany where production is cheaper. In 1999, the volume of sales amounted 226,8 million EUR [KLOOS 2000]. (Presently, China is major supplier of leather goods with a total value of produced goods of 1,12 billion EUR in 1999). Due to work safety reasons, a substitution of the adhesive systems especially by watery adhesive dispersions has taken place over the last years. Substitution was carried out in order to avoid investment needs for installations for extracting exhaust air. The sector has not been investigated.

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5 Wood and Furniture Industry

The application of solvent-based adhesives has been substituted far-reaching by solvent-free products in the industrial wood material and furniture production in Germany. The state-of-the-art is described in the following.

5.1 Production of Wood Materials

5.1.1 General Information

Subject of this chapter is the application of adhesives in wood material production. Wood materials are boards made of wood. [DUBE 1999]. The most important materials for furniture production are:

- Chipboard
- plywood
- beaverboard
- MDF boards (Medium Density Fiberboard).

Among these, chipboard is the most important material in furniture production. Besides the use in the furniture industry, chipboards are used in the construction industry in outside areas as well as inside areas. If a higher mechanical stress is in effect on the material, plywood is used instead of chipboard. Beaverboard is used as back wall of boxes and drawers in furniture construction. As the sector uses big amounts of adhesives, the applied techniques will be described in order to give a documentation of the German state-of-the-art. In Germany, no solvent containing adhesives are processed in the production of wood materials.

In 1998, 8.75 million m^3 of chipboard [HVDH 2000], 407.450 m^3 of plywood and 2,4 million m^3 of beaverboard were produced in Germany. In 1999, there were 134 enterprises with 20.203 employees working in the production of wood material in Germany [SBW 1999].

5.1.2 Applied Technical Processes and Methods

Solvent-free adhesives belonging to the group of polyaddition, polymerization and polycondensation adhesives are processed in the wood material production. As polycondensation adhesives mostly used are synthetic resins based on urea formaldehyde

(UF), melamine urea formaldehyde (MUF), phenol formaldehyde (PF) and polyurethane dimethyl methane diisocyanate (PMDI). These adhesive systems are partly used as adhesive mixtures as well.

If **resins containing formaldehyde** are applied, this might cause formaldehyde emissions due to incomplete condensation of the formaldehyde together with the resin compounds of the adhesives [DUBE 1999, ZEPPENFELD 1991]. For reduction of formaldehyde emissions, different methods are used. Certain substances such as urea, melamine or dicyandiamide may be added as formaldehyde catcher. Formaldehyde that was not used for hardening of the adhesive can be bonded by addition of ammonia (it forms hexamethyl ethylene tetraamine) [DUBE 1999]. The formaldehyde of the adhesive is not a solvent, because it does not assist in processing, but is converted during the chemical reaction.

Adhesive dispersions based on polyvinyl acetate are used for the plywood production in small proportions. Polyvinyl acetate dispersions contain water as dispersing agent.
Small amounts of solvents are contained in adhesive dispersions (e.g. ethyl acetate, aromatics, ketones, alcoholes) to reduce the minimum film temperature and to enhance the contraction of additives. The proportion is typically below 1 % (by weight), but it might be up to 6 %. [SBW 1999]. Due to this low solvent proportion, polyvinyl acetate dispersions can be classified as low in solvents.
The production of wood materials is carried out with solvent-free adhesives almost completely. The applied adhesive techniques are illustrated in the following.

Production of chipboard

Chipboard is the most important material in furniture production. Chipboard is a plate-shaped wood material that is produced by moulding and cementing of wooden parts or wood-like fibres. Chipboard is cemented with chemical reactive solvent-free polycondensates and polymers (polyisocyanate).
Chipboard usually has a three-layered structure. The top layer consists of fine chips with a higher adhesive proportion to form a closed surface. The middle layer consists of coarser particles.

Chipboard for inside use (inside climate conditions) are produced with synthetic resins based on urea or urea melamine formaldehyde resins (UF or MUF resins), because the requirements concerning moisture resistance are lower in this case [ZEPPENFELD 1991].
Chipboard for use in outside areas (outside climate conditions) that requires a higher moisture resistance is produced with phenol resins based on phenol formaldehyde resins (PF

resins). There are also (solvent-free) poly-isocyanates as a formaldehyde-free alternative to PF resins used for this application.

The cited adhesives are applied individually as well as in mixtures. Phenol formaldehyde resins (PF resins) or isocyanates are applied if a higher moisture resistance is required [SBW 1999].

Technically most important adhesives in the chipboard production are the urea formaldehyde resins with a proportion of 85 % [DUBE 1999]. The applied synthetic resins can be adjusted to technical parameters such as moisture of the chips, moulding time and moulding temperature. This is carried out by the selection of the curing agent and its quantity [BÖHME 1975].

Beaverboard

Wood residues, barks and annual plants with a high fibre proportion are raw materials for the production of beaverboard. The components being processed are plastified with steam and formed into boards with a hot press. The process of plastifying releases binders originating from the wood itself, so that the production of these boards is possible without addition of adhesives. The use of adhesives depends on the used raw materials. Adhesives used for beaverboard and plywood for outside areas are PF resins, whereas for inside use UF and MUF resins are applied [BÖHME 1975].

Medium Density Fibreboard (MDF)

Wood chips are raw material for the production of MDF. They are muted under pressure and high temperature and split into fibres. Resins are added to this material. Afterwards, mats are created and pressed. PF and UF resins are used in this process [SBW 1999].

Plywood

Plywood is produced with three veneer wood layers by adhesive processes. The directions of the fibres have to cross with an ankle of 90°. The veneer strips are glued at the joints with UF and PF resins.

For cementing of the middle layer of consisting of rods, partly polyvinyl acetate adhesive dispersions are used besides urea resins [BÖHME 1975]. The outer layers of the plywood are cemented with PF, UF or UMF resins according to the technological requirements of the plywood.

High Pressure Laminated Material Plates (HPL)

High pressure laminated material plates consist of hard paper sheetings impregnated with reaction resins (based on phenol resins) that are piled up forming a core layer, of decorating paper and of the finishing clear overlay paper soaked with melamine resin [BEUTH 1989]. HLP are processed for especially stressed surfaces such as table and kitchen boards.

5.1.3 Current Consumption and Emission Values

In the production of wood materials in Germany, solvent-free adhesive systems are processed which are predominantly based on formaldehyde resins. An exception is gluing of middle layers of plywood where partly polyvinyl acetate dispersions are applied. These compounds may contain small amounts of organic solvents.

5.2 Furniture Production

Solvent-based adhesives have been substituted in Germany by adhesive dispersions, hot melts or reactive systems for most applications. Hence only very small amounts of solvents are processed in the furniture producing installations existing in Germany. As however comparably high amounts of adhesives are processed and the production is partly still carried out with solvent-based products in the European foreign countries, the state-of-the-art in Germany will be described in the following.

5.2.1 General Information

Adhesives in the furniture production are especially processed in the industrial production of wooden kitchen furniture, seat furniture (with frameworks predominantly made of wood), wooden office furniture, wooden furniture for shops, wooden furniture for bedrooms, dining and living rooms.

In 1998, 9.42 million of seat furniture with a wooden frame, 6.14 million of wooden office furniture, 25.9 million of wooden kitchen furniture (including wood furniture for kitchen components), 0.52 million of wooden furniture for shops and 22.62 million of wooden furniture for bedrooms, dining and living rooms were produced in Germany.
In 1999, 836 enterprises worked in the furniture production in Germany having 165,000 employees [HVDH 2000].

The furniture production has been studied exemplarily at three enterprises dealing with the production of kitchen furniture, mattresses and production of shelves and cupboards.

5.2.2 Applied Processes and Techniques

Wood materials are partly coated with foils for furniture production in order to confer them certain technical or optical properties. Urea formaldehyde resins, polyvinyl acetate adhesive dispersions (PVAc), acrylates, polyurethane adhesives, epoxy resins, polyethylene, hot melts (EVA) phenol resins and to a very low extent solvent containing neoprene adhesives are processed for surface finishing.

The adhesives used in the furniture production show a big variety due to the different materials being bonded (wood, glass, plastics, cloths, flexible foam materials). The single adhesives cannot be assigned to the furniture types.

For this reason, the description of the applications is not sorted by the work pieces but by the adhesives processed in the furniture production. Solvent-based adhesives have been substituted almost completely by solvent-free systems up to now. They are only processed for a few special applications.

5.2.2.1 Adhesives

One-component Polyvinyl Acetate Adhesive Dispersions (PVAc)

PVAc are the most important adhesives in the furniture production due to technical reasons affecting their application. They can be applied by different application techniques and also cure without heat's impact. The dispersion agent is water. The adhesives are preferred for assembly adhesive bondings of the indoor area and partly for windows [DUBE 1999].

Decorating laminated material plates and edge bondings are predominantly bonded with PVAc onto wood materials. PVAc are applied for veneering works in a low proportion if application of formaldehyde containing glues should be avoided.

If the glue joint shall be resistant against higher temperatures, alcohols, esters and ketones can be added to the adhesive [ZEPPENFELD1991]. The solvents cause an improved film formation due to welling or dissolving the PVAc particles. The solvent content of these adhesive systems is typically lower than 1 % (weight per cent) and up to 6% for special cases.

Two-component Dispersions

Two-component PVAc adhesive dispersions contain cross-linking agents that increase water and heat resistance if the adhesive bonding. As second component, resins (phenol, resorcin, melamine resins or isocyanates) are added to the compounds. The adhesives are processed for adhesive bondings that have to resist moisture stress at kitchen and bathroom furniture.

Ethylene Vinyl Acetate Adhesive Dispersions (EVA)

Ethylene vinyl acetate adhesive dispersions (EVA) are used for lamination of wood materials with PVC foils. The adhesive systems contain water as dispersion agent. The solids content varies between 45 and 60 %.

Aminoplast and Phenoplast Resins

The aminoplasts UF and MF are used for cementing of veneers, decorating foils, priming foils and to a low extent for cementing of laminated material plates onto wood.
The aminoplasts are cheaper than the PVAc adhesive dispersions, show a faster curing and create more temperature resistant bondings [DUBE 1999]. Phenol resins are used for weatherproof adhesive bondings. The resin adhesives can be applied as cold, warm or hot setting adhesives [ZEPPENFELD 1991].

Hot Melts

Hot melts based on ethylene vinyl acetate copolymers, polyuerethanes and polyamides are processed in the furniture production. Hot melts are used for edge cementing of chipwood, for corpus bondings as well as for application of edge material based on thermoset materials such as polyester edges and melamine resin edges. Hot melts are also used for assembly adhesive bondings and for veneering of large surface areas [DUBE 1999, ZEPPENFELD 1991].

Hot melts are also used increasingly for flexible foam bondings of upholstery instead of solvent-based adhesives. Hot melts used for this process are thermoplastic caoutchoucs, ethylene vinyl acetate copolymers, polyester and polyamide (EVA) [ADHÄSION 2000].
The heat stability of hot melts is up to about 80 °C. Hot melts show a good adhesion with high film strength and a good stability also under low temperature conditions. Due to the fast curing while cooling down, an application of hot melts allows short cycle times. Hot melts are processed as granulates, in cartridges or as powder [ZEPPENFELD 1991].

Neoprene Solvent-based Adhesives

Neoprene adhesives are processed as solution adhesives or adhesive dispersions. Solution adhesives can be used for cementing of wood onto varnished surfaces, as the solution adhesive partly dissolves the varnish surface and therefore creates a good adhesiveness. The solids content of the solution adhesive amounts to 10 to 20 %.
As solvents for the adhesives, mixtures of toluol, xylol, acetone, methyl acetate, ethyl acetate, butyl acetate and cyclohexane are used for technical reasons [DUBE 1999]. Nowadays these compounds are predominantly used in the handcraft section.

Polyurethane Solvent-based Adhesives (PUR)

Polyurethane adhesives are processed with a curing agent based on isocyanates in order to create an additional cross-linking that improves the adhesive strength. PUR adhesives are processed as adhesives for sheating PVC foils on furniture profiles or enclosures (e.g. for loudspeakers) [DUBE 1999].

Styrene Butadiene Styrene Adhesives (SBS)

Solvent-based SBS adhesives (styrene caoutchouc adhesives), hot melts and adhesive dispersions are used for cementing of cushion materials in the upholstery production. Application of these adhesives decreases strongly [ADHÄSION 2000].

5.2.2.2 Adhesive Application

Manual and automated application techniques are used for adhesive application. Manual application techniques include adhesive application with paintbrush, spatula, spray gun as well as dip coating. Casting processes, spraying and dip coating as well as spreading are automated techniques [ZEPPENFELD 1991].

5.2.2.3 Production of Mattresses

In the past, large amounts of solvent-based adhesives were used for cementing of foam layers of different hardness, for cementing of foam elements for innerspring mattresses and for the production of extra-long mattresses. Therefore, the production process will be considered in detail in the following. In Germany, mattresses are produced without any uses of solvent-based adhesives and foaming agents nowadays. In the following, the techniques used in Germany will be described as a documentation of the state-of-the-art.

In 1999, about 1.52 million mattresses were produced by 85 enterprises in Germany.

Applied Processes and Techniques (Dunlopp Company, production site Hanau)

Mattresses are produced out of latex of PU foams. If latex is used, the material consists of synthetic latex predominantly and contains up to 15% natural latex (even more with certain prerequisites) [VWI Verband 2001, LAUBACH 2001].

Polyurethane foams are produced out of polyoles and isocyanates according to the isocyanate polyaddition technique. Depending on the used raw materials, they can be classified as polyether polyoles and polyester polyols. Especially polyether polyols are used in the mattress production.

The foaming operation of the materials is carried out without CFC s exclusively (methylene chloride) in Germany. Latex foams are foamed with air in mixing installations.

Production of PU Mattresses

The fluid material is poured into a cooled, closable mould. The foamed material is created by the development of carbon dioxide during the reaction of isocyanates with water. A foam-like, high molecular polyurethane is created out of polyesters containing diisocyanates on addition of water while carbon dioxide is split off. Shaped pieces are cut out of foam blocks created in the mould for example for seat furniture or mattress production. Individual foam blocks are cemented with solvent-free hot melts.

Production of Latex Mattresses

These mattresses are created by foaming of vulcanizable latex mixtures by gellation of the foam followed by a vulcanisation. For that, 60% latex is mixed with a catalyst, sulfur, an antioxidant, soap and further components depending on the recipe. It is foamed up to the manifold volume and zinc oxide is added as gelling agent. The foaming operation is carried out in either in discontinuous mixers or in continuously working foam machines. The liquid foam is put into a mould and coagulates within 5 to 10 minutes. After closing the mould, it is heated over steam or by high frequency heating in order to vulcanise. After the vulcanisation process, the mattress is washed and dried. After cooling of the mould, a water based mould release agent (wax) is appliquéd onto the inside and the process is repeated.

As only mattresses of standardized sizes can be produced in the industrial production, exceptional sizes are cut off a few foam parts and newly cemented. Hot melts for cementing of mattress materials are based on amorphous polyalphaolefines (APAO) or SBS [ADHÄSION 2000, VWI VERBAND 2001, LAUBACH 2001]. In the innerspring mattress production, the single components are joined by adhesive dispersions. Nowadays no VOC containing products are used any more in the whole mattress production process [VWI VERBAND 2001, LAUBACH 2001].

The production process of latex mattresses is illustrated in figure 5-1. The latex material is mixed in the preparation container, then foamed in the mixer with air in the following processing step and then filled into the heated mattress mould. Here the gellation and vulcanisation is carried out. After withdrawal of the mould, the mattress is eased of the mould release agent by washing and is dried. Solvent-free latex or polyurethane adhesives are applied for the correction of imperfections in very small amounts [LAUBACH 2001].

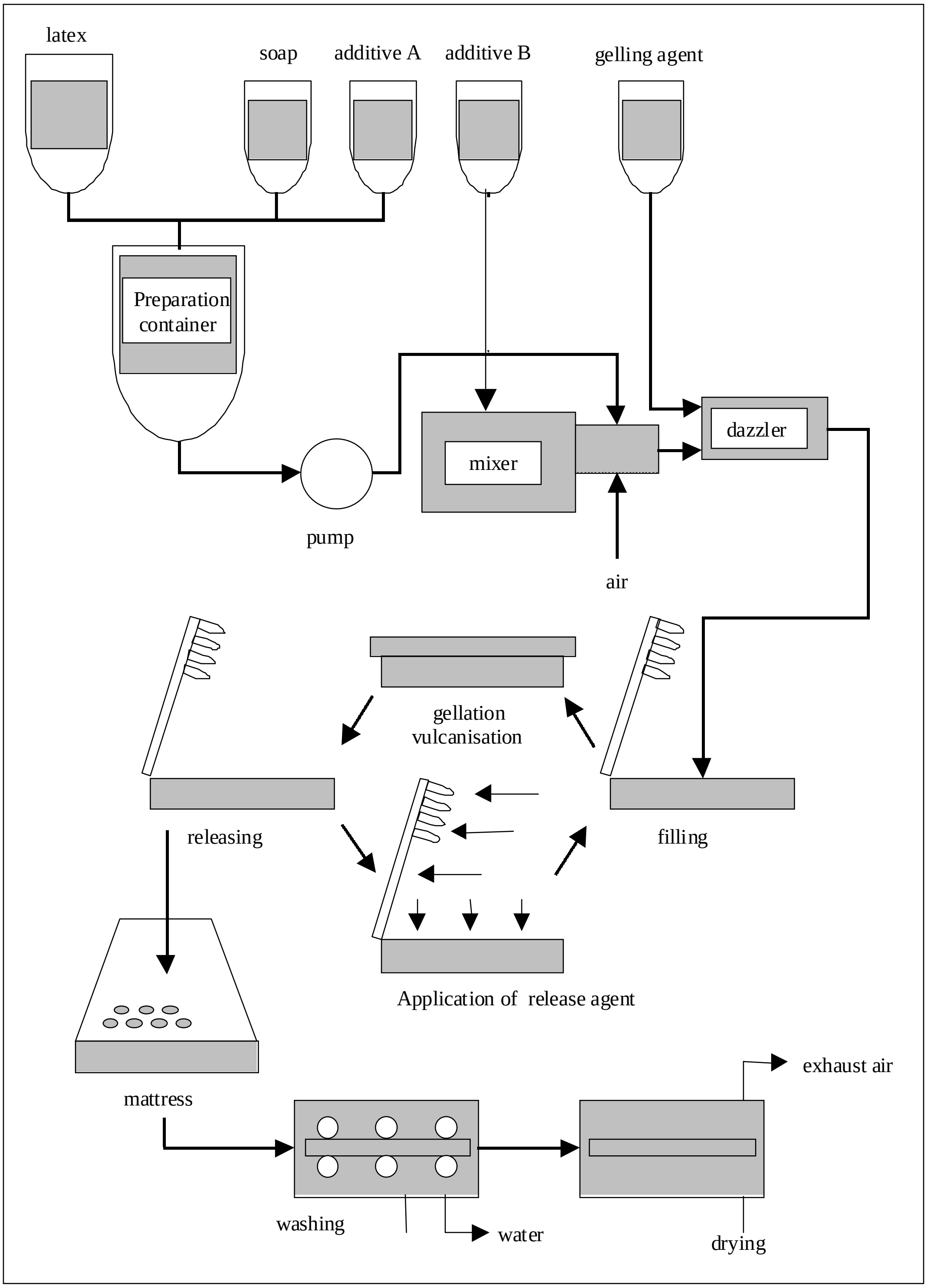

Figure 5-1: *Production of latex mattresses by the Dunlopp technique*

5.2.2.4 Production of cupboards and shelves

The production of cupboards and shelves has been studied at the company Schiedermöbel Wortmann GmbH. In the production (e.g. racks, show-cases, cupboards, mounting furniture), solvent-free adhesives are processed exclusively. The main applications of the adhesives are laminations of chipboards and MDF plates with veneers or decorating foils. Mainly mechanical fixings are used for furniture assembly. To a very low extent, solvent-free cold glues based on urea are applied in the assembly process.

For the plane lamination of wood materials with veneers or decorating foils, urea formaldehyde glues are applied. Lamination is carried out automated. The adhesive is applied onto the surface of the wood materials by roller coaters. The application mass of the glues amounts to about 5 g/m^2 for foil lamination and about 20 g/m^2 for cementing of veneers. The installations are cleaned with water. Narrow edges are coated with foils or veneers in automated processes via with hot melts [SCHIEDER 2001].

5.2.2.5 Production of Kitchen and Bathroom Furniture

The production of kitchen furniture has been investigated at the company ALNO AG in Pfullendorf. Similarly to the production of cupboards etc., solvent-based adhesives are not applied any more nowadays [VWI 2001]. Cementation of foils onto wood materials is carried out with adhesive dispersions based on PUR. These adhesives are applied automatically by compressed air spraying. Before cementing, the adhesives are activated by heat. Solvent-free hot melts are used for cementing of edges.

Producers purchase kitchen work disks from component supplier enterprises. In the production of these work disks, solvent-free resins based on urea formaldehyde are processed.

5.2.3 Current Consumption and Emission Values

The sectors mattress production, cupboard and shelves production and production of kitchen furniture have been studied due to the variety of manufactured products. Solvent containing adhesives are not used in any one of these sectors to a greater extent.

5.2.4 Emission Abatement Measures

The sectors that have been investigated do not show any potential for emission abatement of VOCs with respect to the use of adhesives. The techniques used by the industry can be classified as established BAT.

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6 Industrial Assembly Processes

Assembly processes with adhesive application can be found in the sectors machine construction and electrical engineering.

6.1 Electrical Engineering

Adhesive processes as material sparing joining techniques have a special significance in electrical engineering due to the increasing miniaturization. Adhesive applications can be classified as fixation bondings and contact bondings. Fixation bonding serves for the attachment of components onto printed circuit boards. Electronic components are fixed mostly with UV cross-linking adhesives [INDUSTRIEVERBAND KLEBSTOFFE 2001]. Soldering has often been replaced by contact bonding adhesive applications due to the use of isotropic and anisotropic conducting adhesives. Conducting adhesives are based on UV curing or hot setting epoxy resins, silicones, acrylates and polyamides on addition of conducting particles.

The adhesives used in electrical engineering are solvent-free systems [KLEBEN/DICHTEN 4/2000, 2/2000]. Predominantly UV cross-linking epoxy resins, cyanacrylates, silicones and anaerobic curing compounds are processed [HEßLAND 2001, KLEE 2001], KLEBEN/DICHTEN 11/2000]. According to statements of the producer associations, solvent-based adhesives are not used for the production of electric appliances. Therefore this sector does not fall under the scope of the Directive.

6.2 Machine Construction

Welding and screw driving are the predominant joining techniques in machine construction because especially metallic materials have to be joined. The processed amounts of adhesives are small. Solvent-free adhesive systems such as epoxy resins, cyanacraylates, silicon caoutchouc as well as anaerobic adhesives are used. The sector does not fall under the scope of the Directive [ENDLICH 1997].

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