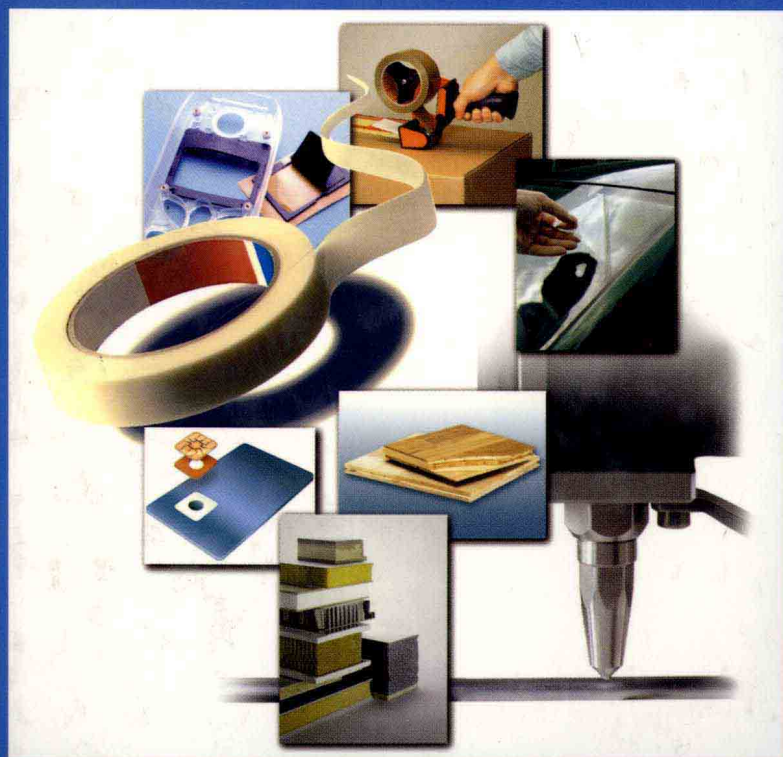
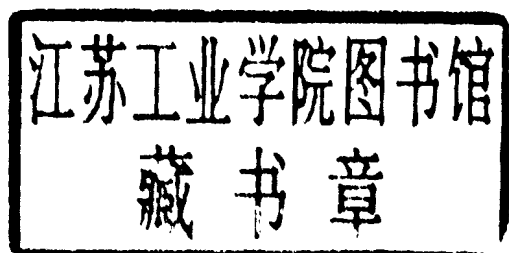


# Adhesives and Adhesive Tapes



Gerhard Gierenz, Werner Karmann (Editors)

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Gerhard Gierenz, Werner Karmann  
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# Preface

The impressive developments in numerous fields of technology – anything we can imagine from bookbinding, food packaging, electronic devices, automobile manufacture to building and aircraft constructions – all engineering progress in these and other areas is enabled and accelerated by innovations in adhesive technology.

Although adhesive applications mostly are not readily evident in daily life – except for some do-it-yourself areas, e. g. paper gluing, and repairs – experts in the development and engineering departments value adhesive technology as an important tool to find solutions to a wide range of technical problems. Strong points are the great variability in the kind and the properties of the adhesives, e. g. drying or reacting liquids, melting solids, adhesive tapes and die cuts.

This book, a special edition of the chapters “Adhesives” and “Adhesive Tapes” of Ullmann’s Encyclopedia of Industrial Chemistry, is intended as a guidance for everybody interested in getting an overview on the various adhesives, their different properties, and the wide field of applications.

Gerhard Gierenz

Werner Karmann

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Gerhard Gierenz

Werner Karmann

# Contents

<b>1. Adhesives . . . . .</b>	<b>1</b>	<b>1.9.7. Bonding of Plastics . . . . .</b>	<b>64</b>
<b>1.1. Introduction. . . . .</b>	<b>1</b>	<b>1.9.8. Bonding of Elastomers. . . . .</b>	<b>66</b>
<b>1.2. Adhesion Theories . . . . .</b>	<b>3</b>	<b>1.9.8.1. Adhesion of Rubber . . . . .</b>	<b>66</b>
<b>1.3. Definitions. . . . .</b>	<b>6</b>	<b>1.9.8.2. Bonding of Rubber. . . . .</b>	<b>67</b>
<b>1.4. Raw Materials for Adhesives. . . . .</b>	<b>10</b>	<b>1.9.9. Bonding of Metals . . . . .</b>	<b>68</b>
<b>1.5. Classification of Adhesives . . . . .</b>	<b>16</b>	<b>1.9.10. Adhesives for Wallcoverings . . . . .</b>	<b>70</b>
<b>1.6. Individual Adhesive Systems. . . . .</b>	<b>19</b>	<b>1.9.11. Floorcovering Adhesives . . . . .</b>	<b>71</b>
1.6.1. Adhesives That Set Without a Chemical Reaction . . . . .	19	<b>1.9.12. Building Construction Adhesives . . . . .</b>	<b>73</b>
1.6.1.1. Solvent-Free Adhesive Systems . . . . .	19	<b>1.9.13. Adhesives for Bonding Textile Fabrics. . . . .</b>	<b>74</b>
1.6.1.2. Adhesive Solutions from Which the Solvents Escape before Bonding . . . . .	23	<b>1.9.14. Flocking Adhesives. . . . .</b>	<b>75</b>
1.6.1.3. Adhesive Solutions from Which the Solvents Evaporate during Bonding . . . . .	25	<b>1.9.15. Adhesives for Bonding Glass . . . . .</b>	<b>76</b>
1.6.1.4. Aqueous Emulsions of Polymeric Compounds. . . . .	28	<b>1.9.16. Adhesives in Automobile Manufacture . . . . .</b>	<b>76</b>
1.6.2. Adhesives Setting by Chemical Reaction. . . . .	29	<b>1.9.17. Adhesives in Aircraft Construction . . . . .</b>	<b>80</b>
1.6.2.1. Adhesives Setting by Polymerization . . . . .	30	<b>1.9.18. Adhesives and Sealants in Electronics . . . . .</b>	<b>83</b>
1.6.2.2. Adhesives Setting by Polyaddition . . . . .	35	<b>1.9.19. Medical Adhesives . . . . .</b>	<b>86</b>
1.6.2.3. Adhesives Setting by Polycon- densation . . . . .	38	<b>1.9.20. Household Adhesives . . . . .</b>	<b>89</b>
1.6.2.4. Vulcanizing Adhesives . . . . .	41	<b>1.9.21. Applications of Anaerobic Adhesives. . . . .</b>	<b>93</b>
1.6.2.5. Ultraviolet/Electron Beam (UV/EB) Curing Adhesives . . . . .	41	<b>1.10. Economic Aspects . . . . .</b>	<b>94</b>
1.6.2.6. Conductive Adhesives . . . . .	43	<b>2. Adhesive Tapes . . . . .</b>	<b>97</b>
<b>1.7. Bonding Techniques . . . . .</b>	<b>44</b>	<b>2.1. Introduction. . . . .</b>	<b>97</b>
<b>1.8. Testing of Adhesives . . . . .</b>	<b>49</b>	<b>2.2. Pressure-Sensitive Adhesion . . . . .</b>	<b>98</b>
<b>1.9. Applications of Adhesives . . . . .</b>	<b>52</b>	<b>2.2.1. Mechanism of Pressure-Sensitive Adhesion . . . . .</b>	<b>99</b>
1.9.1. Bookbinding . . . . .	52	<b>2.2.2. Dynamic Mechanical Analysis . . . . .</b>	<b>101</b>
1.9.2. Adhesives for Packaging and Packaging Materials . . . . .	54	<b>2.3. Test Methods . . . . .</b>	<b>101</b>
1.9.3. Nonwovens Hygiene Industry. . . . .	57	<b>2.4. Raw Materials. . . . .</b>	<b>104</b>
1.9.4. Hygienic Papers . . . . .	58	<b>2.4.1. Elastomers . . . . .</b>	<b>104</b>
1.9.5. Gluing of Wood and Wooden Materials . . . . .	58	<b>2.4.2. Tackifier Resins. . . . .</b>	<b>106</b>
1.9.6. Footwear Adhesives . . . . .	62	<b>2.4.3. Plasticizers . . . . .</b>	<b>108</b>
		<b>2.4.4. Fillers . . . . .</b>	<b>108</b>
		<b>2.4.5. Stabilizers. . . . .</b>	<b>109</b>
		<b>2.4.6. Release Coatings . . . . .</b>	<b>109</b>
		<b>2.5. Manufacturing Processes . . . . .</b>	<b>110</b>
		<b>2.5.1. Compounding . . . . .</b>	<b>112</b>
		<b>2.5.2. Coating . . . . .</b>	<b>112</b>
		<b>2.5.3. Drying. . . . .</b>	<b>113</b>



2.5.4.	Cross-linking / Curing . . . . .	114	2.6.1.6.	Electrical Tapes . . . . .	120
2.5.5.	Slitting, Cutting . . . . .	115	2.6.2.	Double-Sided Adhesive Tapes. .	121
<b>2.6.</b>	<b>Adhesive Tape Products. . .</b>	<b>116</b>	2.6.2.1.	Standard Double-Sided Tapes. .	121
2.6.1.	One-Sided Adhesive Tapes . . .	116	2.6.2.2.	Transfer Tapes . . . . .	122
2.6.1.1.	Film Tapes . . . . .	116	2.6.2.3.	Adhesive Pads and Strips . . . .	123
2.6.1.2.	Fiber-Reinforced Tapes. . . . .	119	2.6.2.4.	High-Performance Tapes. . . . .	123
2.6.1.3.	Paper Tapes. . . . .	119	2.6.3.	Medical Tapes . . . . .	123
2.6.1.4.	Textile Tapes . . . . .	119	<b>2.7.</b>	<b>References . . . . .</b>	<b>125</b>
2.6.1.5.	Other Backings . . . . .	120	<b>Subject Index . . . . .</b>	<b>135</b>	

# I. Adhesives

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## I.1. Introduction

Adhesives are defined as nonmetallic substances capable of joining materials by surface bonding (adhesion), the bond itself possessing adequate internal strength (cohesion). Adhesive is a generic term and covers other common terms, such as glue, paste, gums, adhesive cement, and bonding agent.

**History.** Adhesion is among the oldest technologies of mankind [129]. As early as the Stone Age, our ancestors made stone axes and other tools using mineral pitch or wood rosin. Asphalt was used in building the Tower of Babel. In ancient Egypt, it was already customary to veneer furniture with the aid of animal glues. Flour- and casein-based adhesives were applied in ancient Rome. Adhesives of plant origin such as wood rosin were as well-known in China, as was the gum arabic and the caoutchouc (“weeping wood”) from the tropical regions of South America and Asia.

In the Middle Ages, the first glue-boiling plants came into being and produced protein glues from animal raw materials (glutin from hides and bones, blood albumin, casein from milk) or starch paste from plants.

With the developments in chemistry in the early 1900s, the technology of adhesives began to boom. Phenolic resins (ca. 1900), melamine resins, and urea resins (ca. 1930), polymer dispersions, epoxy resins (1938), and cyanoacrylates (1957) largely superseded the classic natural adhesives and, with a multitude of bindings agents, form the basis for modern adhesive technology, which is one of the most advanced joining processes.

**Composition.** An adhesive is composed of basic raw materials, which are called binders [1] and which determine its adhesiveness (adhesion) and its internal strength (cohesion), and of frequently necessary auxiliaries, which establish particular end-use and processing characteristics. The adhesiveness of an adhesive, its internal strength after setting, and its processing characteristics are the fundamental properties that determine its suitability for use in forming adhesive joints. Adhesive joints are the joints formed between substrates and adherents using adhesives.

The binders used for adhesives are primarily high polymers having optimal strength properties. High internal strength (cohesion) is essential if the adhesive in an adhesive joint is to be able to transmit forces from one adherent to the other. Most adhesives contain high molecular mass organic substances as their basic raw materials or reactive organic compounds that are preliminary stages of polymers and that react during the bonding process to form polymers. Inorganic polymers, such as the various types of waterglass, are used only to a very limited extent.

Virtually any standard polycondensate, homopolymer, and copolymer and also polyadducts may be used, provided they can be applied as solutions, dispersions, emulsions, or melts. In addition to these raw materials, auxiliaries such as resins, plasticizers, fillers, thickeners, solvents, antiagers, preservatives, hardeners, or setting retarders, are required, depending on the end use. Their function is inter alia to adjust tack, to improve adhesion, to make flexible, to regulate viscosity, to stabilize, and to influence setting or hardening.

**Adhesive Joints.** In many cases, adhesion is as effective as other joining techniques, such as riveting, welding, soldering, and screwing; in some cases it complements those techniques; and frequently it affords numerous advantages. On the other hand, certain processing requirements and material properties related to chemical structure can restrict the use of adhesives.

Bonding has the advantage that the joining of adherents can be carried out very rationally, often extremely quickly, and particularly economically [2]. One important feature common to all adhesive joints is the highly uniform distribution of forces over the entire joint area by comparison with rivet and screw joints. The uniform stress levels frequently provide for more optimal utilization of material strength. Accordingly, thinner and hence lighter sections of material may be used. This has resulted in the saving of material and, for example, in the automotive, aircraft, manufacturing, and furniture industries, in totally new structural elements (sandwich elements).

The use of adhesives enables numerous materials, even those differing widely in type, to be joined to one another. This applies above all to materials that cannot be joined to one another by other techniques. Because some adhesives set even in relatively thick layers, any unevenness in the constituent material of the adherents may be smoothed out during bonding, or significant dimensional tolerances may even be bridged by the gap-filling principle. The adhesive layer of an adhesive joint even may have a vibration-dampening effect and, by virtue of its insulating properties, can prevent contact corrosion in adhesive joints involving metals of different normal potential. Another advantage is the fact that many adhesives can be applied at room temperature or, when heat must be used for application, it is sufficient to apply temperatures at which the constituent materials of the adherents are not affected, as can happen in the welding of metals and plastics.

The usability of adhesives can be restricted when stringent requirements are imposed on the thermal stability of an adhesive joint [2]. Like all plastics, adhesives based on organic polymers also show a marked dependence on temperature in their strength properties. The strength properties of an adhesive joint can be kept constant and sufficiently high in only a relatively narrow temperature range. Adhesives cured by thermal cross-linking which are still relatively temperature resistant are generally capable of withstanding temperatures of up to about 150 °C. Using new, but more difficult to apply adhesives based on polyimides [3], it is possible to form adhesive joints which are capable of withstanding temperatures of the order of 250 °C for long periods. On the other hand, some adhesives, when subjected to heavy static stress for long periods, show a tendency even at room temperature to creep, a plastic deformation that can result in separation of the adhesive joint.

## 1.2. Adhesion Theories

The atoms or molecules in any solid are held together by so-called cohesion forces. These interatomic and intermolecular forces are conservative in character, i.e., not dependent upon the pretreatment of the material [4, p. 1]. After a break in the solid, the two fragments cannot be put back together in such a way as to restore the solid to its original state. Reactivation of the cohesion forces would be possible only if the original intervals between atoms or molecules could be reestablished. The same may be

said also of the joining of two different materials; a mutual convergence over the entire joint area on the radius of interaction of the molecules would result in adhesion of the same order of magnitude as the cohesion forces [5]. In practice such convergence is not possible. There are three reasons for this: first, the surface roughness of the adherents; second, a reduction in the effective surface for boundary layer reactions due to the surface tension of the adhesive; and third, the formation of the so-called weak boundary layer [130], [131]. This is a region within the adhesive film close to the boundary layer where the reactive groups of the molecules are directed to the adherent and consequently do not contribute to the curing process, that is, no cross-linking occurs. Therefore, the weak boundary layer is always a weak link in the adhesive joint. The function of the adhesive in the boundary zone is to compensate for the effects of surface roughness by providing optimal wetting of the adherents.

Over the years, several different theories have been developed as to the mode of action of adhesives, i.e., the mechanism of adhesion. They extend from the simple mechanical theory via the electrostatic and adsorption theory to the diffusion theory, these being the most important [6]. One feature common to all these theories is that, for some adhesion phenomena, these theories allow qualitatively satisfactory assertions to be made, but in other respects they fail totally and, in some cases, even lead to conflicting results. The adhesion of adhesive to an adherent, on which any joint is based, is clearly not a consistent and isolatable process [7] but a complex addition of various adhesion effects. Because the number of individual processes involved, apart from exceptions, is extremely difficult to estimate, definitive confirmation or rejection of individual theories is hardly possible.

**Mechanical Theory of Adhesion.** The oldest theory of adhesion is definitely the mechanical theory. It is based on mechanical anchorage of the adhesive in pores and irregularities in the adherent and is discussed primarily in reference to wood and similar porous materials [8].

**Electrostatic Theory of Adhesion.** According to the electrostatic theory [9], the adhesion forces between adherent and adhesive layer are applied by contact or transfer potentials. These transfer potentials cause the buildup of an electric double layer at the adhesive–adherent boundary and corresponding Coulomb attraction forces between the two components. In principle, the occurrence of transfer potentials is unquestionable, as shown by the electrostatic discharges that can be detected in the destruction of adhesive joints. However, the practical significance of the attraction forces associated with these discharges is still being debated [4, pp. 150–153], [10].

**Adsorption Theory.** As its name indicates, this theory of adhesion draws upon surface forces for explaining the observed phenomena [11]. It regards adhesion as essentially a special property of phase interfaces. The forces that are responsible for adhesion in this process are the so-called secondary valence or van der Waal's forces. These forces have three components, namely Keesom's dipole orienting effect, Debye's

induced dipole effect, and London's dispersion effect [12]–[14]. For these forces to become active, the distances between the molecules of adhesive and adherent must converge toward molecular intervals. This requires complete spreading of the adhesive over the surface of the adherent. Good adhesion can be expected if the adhesive (in liquid form or in a liquid medium) wets the adherent [8]. Accordingly, the adsorption theory may be placed on an entirely thermodynamic basis. Adhesion is thus determined by the ratio between the surface energies of the adhesive and the constituent material of the adherent in the sense that the specific surface energy of the adhesive must be lower than that of the adherent. Accordingly, materials having high surface energy levels, such as metals, and those having medium surface energy levels, such as wood and paper, may be bonded relatively easily. In the case of polymers, bonding becomes increasingly more difficult [15], and finally almost impossible as surface energy decreases (polyolefins and polytetrafluoroethylene). The remedial surface treatment of materials such as these may be interpreted as increasing their surface energy [16].

The adsorption theory also shows that the adhesion forces of two materials are not reciprocal. For example, if a liquid epoxy resin adhesive is allowed to set on the surface of polytetrafluoroethylene or polyethylene, a very weak adhesive joint is formed. If, by contrast, liquid polytetrafluoroethylene or polyethylene is applied to the surface of hardened epoxy resin, strong adhesive joints are obtained [17]. In practice, this theory is not free from contradictions either; above all, it does not answer the question whether the difference in the surface energies between two materials is indicative of the intensity of the adhesion force [18].

**Diffusion Theory.** The diffusion theory of adhesion is essentially applicable to the bonding of high polymers. According to this principle, adhesion is obtained by the mutual penetration of adhesive and substrate [19], [20]. This mobility is based on the fundamental properties of high polymers: their chainlike structure and resulting mobility, allowing the possibility that the chains possess Brownian molecular movements in a submolecular range. By virtue of their greater mobility, the adhesive molecules normally play a greater part in the diffusion process. However, if the adhesive is in solution, which is generally the case, and if the substrate is slightly soluble in the solvent, substrate molecules or parts thereof also diffuse into the spread adhesive. A diffusion bond is characterized by the disappearance of a clear boundary between the two phases and by the development of a gradual transition from one phase to the other. The mechanism of adhesion has developed into a three-dimensional process and is no longer confined to one interface.

The interdiffusion of the polymer molecules of adhesive and substrate is dependent upon various parameters, such as pressure, time, temperature, molecule size, and, of course, the reciprocal solubility, as shown by the correlation between the compatibility of the polymers and the quality of the bond [21]. Examples of bonds to which the diffusion theory is applicable include the bonding of PVC-U adherents to PVC in solvents containing tetrahydrofuran and so-called contact bonding, where the diffusion

process takes place between two adhesive layers and not between adhesive and substrate [22].

The limits of the diffusion theory of adhesion show up in the adhesion of polymers, for example, to metal or glass. In this case, this theory does not make any useful contribution to the understanding of adhesion.

**Other Theories.** In addition to these theories, some special cases are discussed in the literature. They include *adhesion by primary valence forces*, for example, in the bonding of metals [8], [23], and so-called *liquid adhesion*. In the latter, a thin film of a liquid of extremely high viscosity produces adhesion through a process in which separation of adherent and substrate results in a flow in the narrow gap which is only possible by overcoming considerable resistance. Liquid adhesion is particularly assumed in pressure-sensitive bonding [16] and in the initial tack of a liquid adhesive.

**Summary.** None of the theories covers every single aspect of bonding. A promising combination of the theories to bring about an improvement does not exist at present. In addition, the theory of adhesion as a physicochemical phenomenon and physically measurable adhesion are generally still a considerable distance apart, so that the adhesion theories are only reference points. Secondary factors, such as surface roughness, boundary layers with faults (atmosphere, impurities), joint design, type of stress, and aging also influence bond strength to a considerable extent.

## 1.3. Definitions

Some of the terms are defined in DIN 16920 [24] and EN 923 [132].

**Adherent:** a body that is or is intended to be bonded to another body.

**Adhesion:** state in which two surfaces are held together by interfacial bonds.

**Adhesive application:** the application and distribution of an adhesive over the surface to be joined.

**Adhesive coat:** adhesive layer applied to an adherent.

**Appearance of separated surfaces:** surfaces formed in the event of failure of an adhesive joint; they provide information on the reason for the failure. Distinctions can be drawn visually between:

- 1) A bond layer that does not cover the entire joint area (uneven adhesive application)
- 2) Adhesion failure: rupture of the bond layer from the surface of the adherent
- 3) Cohesion failure: break in the bond layer

- 4) Insufficient coalescence: separation of two adhesive layers at the interface, for example, in the case of contact adhesives
- 5) Rupture of the surface layer of an adherent, breakage of the adherent (material failure)

In many cases, several types of failure occur together.

**Assembly time:** interval between application of adhesive to the adherents and the initiation by heat and/or pressure of the setting process in the assembled joint.

**Application weight, spread of adhesive, coverage:** quantity of adhesive applied per unit area of a surface. A distinction is drawn between wet application (including solvent or dispersant) and dry application (100 % adhesive). Each type of adhesive has technically and economically an optimal application weight. In principle, rough and porous substrates (paper, fabrics) require more adhesive than smooth surfaces (glass, metals, films) to obtain whole-surface wetting and bonding. Application weights range from  $1 \text{ g/m}^2$  (for example, for bonding films) to several  $100 \text{ g/m}^2$ , depending on the substrate, the method of application, and the type of adhesive.

**Bond layer thickness:** the thickness of the set layer of adhesive in millimeters. It may be influenced by the application weight and the fixing pressure. In general, a thin ( $< 0.001 \text{ mm}$ ) and uniform bond layer leads to the highest bond strengths.

**Bond line:** adhesive layer in a bonded joint.

**Bond strength:** force necessary to bring an adhesive joint to the point of failure in or near the plane of the bond line.

**Closed assembly time:** interval between assembly of the adhesive joint and the initiation by heat and/or pressure of the setting process in the assembled joint.

**Cohesion:** state in which the particles of a single substance are held together by primary or secondary valence forces.

**Cold flow:** deformation of an adhesive layer (or film) at room temperature without external applied load.

**Creep:** slow inelastic deformation of an adhesive layer under load following initial instantaneous elastic or rapid deformation.

**Curing:** the setting of an adhesive by chemical reaction (polymerization, polycondensation, polyaddition).



**Heat resistance:** the maximum temperature that a set adhesive in a joint is capable of withstanding. It depends upon the nature and intensity of the load and must be determined in each individual case.

**Initial tack:** the ability of a setting adhesive to hold two substrates together immediately after they have been joined. At this stage of the bonding process, strength is determined largely by rheological properties and depends on the cohesion of the unset adhesive. In the case of hot-melt adhesives, the analogous term “hot tack” is used, and this property is determined by the viscosity of the unsolidified melt. The transitions between the use of the terms initial tack and tack are fluent.

**Minimum film-forming temperature:** the temperature at which an emulsion-based adhesive just no longer dries to form a clear, homogeneous film.

**Open assembly time, open time:** interval between adhesive application to the adherents and assembly of the adhesive joint. The expression “open time” also is used occasionally for the period of time that elapses in a machine between application of the adhesive and bonding.

**Primers:** (mixtures of) substances that are applied before the adhesive to one or both of the surfaces to be joined, for improving adhesion and/or durability of the bond.

**Setting:** process by which an adhesive develops its cohesive strength and hence the physical and chemical properties of its bond.

**Setting rate:** the increase in strength in the bond line per unit time. It depends upon the setting mechanism and temperature and also may be influenced by the substrate (absorbency), bond layer thickness, etc. The setting rate generally is not constant and decreases toward the end of the setting time.

**Setting time:** time between joining of the adherents and the setting of the adhesive. It varies from a few seconds to several days, depending on the type of adhesive.

**Shear strength:** force per unit surface area necessary to bring an adhesive joint to the point of failure by means of shear forces.

**Shelf life, storage life:** time of storage under stated conditions during which an adhesive can be expected to retain its working properties. Appropriate storage generally is understood to mean that the adhesives are stored in clean, hermetically sealed containers and in cool, dry, well-ventilated rooms. Shelf life is largely dependent upon temperature. Aqueous and in some cases even solvent-containing systems have a tendency for skin formation, separation, and an increase in viscosity, particularly when stored for long periods at relatively high temperatures. They also can be spoiled by