Adhesives and Glues

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Keywords
Adhesives · Glues · Epoxy resins · Acrylates · Colophony · PTBP

1 Core Messages

- Exposure to adhesives and glues is common in occupation, leisure time, and household activities. There is a constantly growing group of occupational glues in which the adhesive is a polymerization product formed by a complex chemical reaction between the macromolecules.
- Phenol-formaldehyde resins have occupational relevance for shoemakers, in car manufacturing, in the wood industries, and in construction industries.
- Epoxy resins, used in a wide range of industries, penetrate regular gloves and may cause aerogenic contact dermatitis.
- While acrylic adhesives are less sensitizing than epoxy and colophony rosin, acrylates present with cross-reactions among each other, but not between methacrylates and cyanoacrylates.
- Colophony rosin is a natural material and also used in many occupational fields.

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S. M. John et al. (eds.), *Kanerva's Occupational Dermatology*,
https://doi.org/10.1007/978-3-319-68617-2_59

- Glues should be tested epicutaneously either using commercial patch-test substances or as is but only after having completely dried in order to minimize the risk of active sensitization via patch testing.

2 Introduction

Exposure to adhesives and glues is common in occupational, leisure, and household activities. Multiple types of glues and adhesives are in use, depending on the materials to be joined. Whereas a century ago only paper, wood, leather, and textiles could be glued, today almost any type of materials can be fixed to each other.

In the majority of glues, the action is simply due to removal of solvents, cooling, or crystallization. In these cases, it is not the action of the macromolecular molecules but rather the preservatives, detergents, and other additives added to the adhesive preparation that may cause dermatological problems, although this is rare. There is, however, a constantly growing second group of glues in which the adhesive is a polymerization product formed by a complex chemical reaction between macromolecules. This second group may cause many more irritant and allergic reactions on the skin (Malten 1984).

In this instance, two-component systems are common that react only after being mixed. Other ways to start the reaction include heating, UV irradiation, oxygen and air exposure, pressure-induced rupture of catalyst reservoirs, etc. The focus is mainly on this second group of adhesive chemicals and their occupational use.

3 Formaldehyde Resins

Phenol-formaldehyde resins resemble a group of chemicals that contain the formaldehyde structure but are not necessarily associated with formaldehyde allergy. Resols and novolacs are distinguished within that group. While the chemical curing of novolacs requires the presence of formaldehyde to react with the phenol terminate group, resols do not. Resols are intermediates, which, in turn, need heat to cure (Malten 1984). Para-substituted phenol resins do not cross-link but adhere readily under pressure. Among them, *para*-tertiary butylphenol-formaldehyde resin (PTBP-FR) is a well-known allergen with particular use as neoprene-based leather glue. Therefore it has occupational relevance for shoemakers. An Italian survey revealed PTBP-FR-containing neoprene adhesives to be the major allergens in a shoe factory (Manusco et al. 1996). Apart from PTBP-FR, mercaptobenzothiazole (MBT), two-component-polyurethane-, and epoxy resin-based glues were also found in the same study as relevant glue allergens with special applications each in shoe manufacture. PTBP-FR-based glues have also been reported for their use in car manufacture (Schubert and Agatha 1979).

Phenol-formaldehyde resin is used, for example, in glass microfiber production as a blend for the fiber to strengthen it (Sripaiboonkij et al. 2009). Exposure of the workers to the coated microfibers occurs via inhaling and direct contact. This leads besides respiratory, nasal, and eye symptoms also to skin symptoms with an exposure-response relation.

Foot dermatitis elicited by shoes and wrist dermatitis by watch straps and by other leather articles is mostly not occupationally relevant (Freeman 1997). Of 839 Finnish patients, 9 patch tested with a glue series reacted to PTBP-FR, which turned out to be the most common relevant glue allergen (Tarvainen 1995).

Despite the similar chemical nature, there is no evidence for cross-reactivity between phenol-formaldehyde resin and PTBP-FR (Geldof et al. 1989). Some authors have attributed test reactions to PTBP-FR to free formaldehyde; however, most patients are actually not allergic to formaldehyde. Schubert and Agatha (1979) performed patch-test studies with the chromatographically extracted ingredients of commercially available PTBP-FR. They found two linear condensates named 2-hydroxy-5-tert-butylbenzylalcohol and 2,2′-dihydroxy-3,3-di-(2-hydroxy-5-tertiary butyl)-benzyl-5,5-ditertiary butyl-diphenylmethan to be the real allergens.

PTBP is used in the production of PTBP-FR. Formerly, PTBP has been added in excess, which might have caused concomitant reactions,

whereas nowadays this is no longer relevant. PTBP and *para*-tertiary butylcatechol, both antioxidants in plastics, might cross-react. As a PTBP-FR, they are also known for their toxic effects on skin. Toxic exposure to PTBP-FR may cause chemical burn and even toxic leukoderma. Vitiligo-like leukoderma on the hands of PTBP-FR-exposed shoe-manufacturing workers has been found in the above-cited Italian study. For patch-test purposes, PTBP-FR is available 1% in petrolatum by all well-known suppliers, either in a special plastic and glue or in the shoe series.

Urea-formaldehyde resin and melamine-formaldehyde resin are used as glues in the wood industries to make furniture press plates. Despite a low constant release of formaldehyde from these plates into the indoor air, the health effect for individuals living or working in the room is way overestimated in our opinion. Construction workers are also exposed to formaldehyde resins in modern building materials. Textile finishes are another use for these formaldehyde resins (Fowler et al. 1992). Even cosmetics may contain PTBP-FR as Angelini and others have shown (Angelini et al. 1993). Both resins are currently available from Chemotechnique, Sweden, urea-FR as a 10% petrolatum and melamine-FR as a 7% preparation in the textile color and finishes series.

If nail varnishes are considered to be adhesives, then toluene-sulfonamide-formaldehyde resin (TS-FR) should be included in this chapter. As a very common ingredient of nail lacquers and hardeners, it may be an occupational allergen for cosmetologists, beauticians, and, in a wider sense, those who use these products and who are in public service, such as bank clerks, office employees, and salespersons. Among patients with cosmetic-related contact dermatitis, TS-FR was found to be relevant in 12.6% of the cases, second only to skin care products (De Groot et al. 1988). Typically, TS-FR allergy involves the eyelids, lateral aspects of the neck, and, more seldom, the truly exposed periungual area. Because traces of the allergen are easily transferred to the eyelids, it has been included in the topical eye preparation series of several allergen providers. Almost all brands of nail polishes contain TS-FR (Hausen et al. 1995; Sainio et al. 1997). TS-FR has recently been included in most cosmetic patch-test trays. In addition to TS-FR, acrylates are gaining more and more importance in nail varnishes and lacquers. Chemotechnique provides a separate acrylic nail test series for this purpose.

4 Epoxy Resins

When used for industrial adhesive purposes, epoxy resins occur mostly in the construction industry; the assembly of cars, ships, and airplanes; the manufacturing of sports equipment; as well as in the optical and electronic industries. Much is similar between the chemistry of epoxy resins and acrylates. Both are (often) two-component systems. They are not sensitizing when fully cured but frequently contain an amount of remaining monomer, which is enough to booster a preexisting sensitization. Both penetrate regular gloves which makes skin protection a real problem, especially for sensitized people.

Epoxy resin monomers are polymer precursor units that are reacted with hardeners to give the polymeric material (Hagvall et al. 2016). Approximately 75–90% of all epoxy resins are polymers of diglycidyl ether of bisphenol A (DGEBA), and 1% of epoxy resins are polymers of diglycidyl ether of bisphenol F (DGEBF). DGEBA and DGEBF are considered to be the major sensitizers. The contact allergenic effect of the epoxy resin monomers depends on the reactive epoxide groups (Hagvall et al. 2016; O'Boyle et al. 2012).

Many different chemicals may be added to improve or adapt the material to the required condition. Hardeners of the amine or acid anhydride type are cross-linking agents for the resin (White 1990). They are usually dissolved in organic solvents. Some of those additional chemicals, such as glycidyl ether, benzol, and toluol, are skin irritants, which may enhance the skin damage and thereby booster the induction or elicitation of an allergic reaction.

Workers in the electronics, optical, paint, and glue industries are most likely to acquire occupational allergy to epoxy resins (Richter 1974; Tosti et al. 1993). A Dutch survey among employees of several companies that specialize in epoxy resin-

related work in the construction industry revealed hand eczema in 23 of 135 persons. Of these, 61% of the eczema population and 12% of the healthy skin group had positive patch-test reactions. Almost all positive patch-test reactions were due to epoxy resin. This shows the tremendous relevance of epoxy resin as occupational allergen (van Putten et al. 1984). Epoxy resin-coated fiberglass fibers have been reported as a cause of dermatitis in this particular field of industry (Holness and Nethercott 1989).

Pesonen et al. (2015) analyzed the pattern of patch-test reactivity to allergens in the European baseline series of patients with occupational contact dermatitis in different occupations. Contact allergy to epoxy resin was most commonly found among floor layers and tile setters, 23.8% of whom were patch-test positive for epoxy resin (Pesonen et al. 2015).

Occupational exposition to epoxy resin should be considered in patients with face dermatitis and proved sensitization to epoxy resin. The German information network of dermatology clinics reported higher rates of sensitization to epoxy resin especially in men with face dermatitis which correlates with the more frequent occupational (aerogenic) contact to epoxy resin, for example, in the production of cars, ships, machines or in metal, chemistry, and construction industries (Schnuch et al. 2009).

There are frequent reports about non-occupational cases of epoxy resin sensitivity too. Contact dermatitis due to the epoxy resin in textile label patches has been attributed to the epoxy-containing adhesive (Fregert and Orsmark 1984). Limiting patch tests with epoxy chemicals to the standardized test series is recommended because of the danger of active sensitization. Once proven, epoxy resins should not be retested, in our opinion. When testing epoxy-exposed individuals, never forget to include chemical additives such as the broad range of hardeners.

Epoxy resins are in discussion to cause or trigger the onset of systemic sclerosis. There was also a description of a disease of skin sclerosis and muscle weakness after occupational exposition to vapor of epoxy resin (Yamakage et al. 1980). Animal and patient studies revealed that the most likely etiological agent in the epoxy resin is cyclohexylamine. Nevertheless, correlations between epoxy resin as a possible causer and sclerotic skin diseases are weak, not at last because of the low frequency of these diseases in comparison to the exposure rate to epoxy resins (Steen 1999).

5 Acrylates

Apart from their use in plastics, colors, lacquers, coatings, dental materials, orthopedic appliances, etc., acrylates are now increasingly used in adhesives (Kanerva and Alanko 1998; Table 1) because of their excellent properties, such as strong adhesion – even to metals, ceramics, glass, and other building materials – fast curing, and easy handling. Stickers, tapes, and office material may also be based on acrylic adhesives. An extensive list of ingredients in pressure-sensitive tapes was published in *Fisher's Contact Dermatitis*. The most common allergenic ingredient of those tapes is 2-ethylhexyl acrylate. Occupational problems can arise from workplaces where stickers and labels are put on the final products or, theoretically, in the medicine. However, compared with epoxy and colophony rosin, which have been preferred in the past, acrylate-based tape adhesives are less sensitizing, and reports are based on single cases. There are one- and two-component acrylic adhesive systems. Epoxy (meth)acrylates are a combination of epoxy resins or its raw materials bisphenol A and epichlorohydrin with acrylates and methacrylates. Positive patch-test results to epoxy (meth) acrylates are often not caused by a specific

Table 1 Adhesives based on acrylates, methacrylates, and epoxy diacrylates according to Kanerva and Alanko (1998)

Anaerobic sealants
Cyanoacrylates
Ultraviolet-cured sealants
Methyl methacrylate
Metal and glass glues
Epoxy diacrylates (vinyl resins)
Acrylic dental bonding agents

occupational exposure to these substances but due to a sensitization to diglycidyl ether of bisphenol A. This has relevance for various occupational fields (Aalto-Korte et al. 2009)

Hazardous health effects on the skin are allergic sensitizations and irritation to several acrylates. Glue-induced adherence of contaminated skin (finger, eyelids) may be very difficult to separate. If possible, wait for spontaneous resolution; gentle teasing with repeated moistening of the skin may be applied in this otherwise harmless condition. Respiratory diseases like asthma, eosinophilic bronchitis, and rhinitis have often been reported when handling acrylics (Savonius et al. 1993; Kopferschmidt-Kubler et al. 1996; Quirce 2004).

As known from other fields of (meth)acrylate use (dental prostheses, daily plastic wear), allergy can only be initiated and elicited by uncured monomers. In contrast to plastic wear, monomers are always available when using glues. So the rate of sensitization could be rather high and similar to that of monomeric acrylate use in other occupational fields, such as dentistry. There is, however, only scanty data about real incidence of acrylate sensitivity caused by acrylic adhesives or glues in occupationally exposed individuals or even those within the normal population. Obviously there is (still) no major problem evolving from non-occupational temporary use of these substances, although several reports on sensitization from an adhesive in an electrosurgical grounding plate were reported (Kanerva and Alanko 1998). In occupational medicine, acrylates are important sensitizers to keep in mind. Sometimes it may be hard to find suitable personal protective equipment for sensitized people because most acrylates do penetrate gloves easily.

Cyanoacrylates are among the most common ingredients of acrylate glues (loctite is a well-known brand example). Eyelid eczema, nummular eczema on the hands, and periungual dermatitis and even allergic onycholysis (Kanerva and Estlander 2000) are features of allergic contact dermatitis caused by cyanoacrylate glue used to fix artificial nails. Ethyl cyanoacrylate and methyl methacrylate were seen as allergens in these patients (Belsito 1987; Guin et al. 1998) as well as 2-hydroxy methacrylate, 2-hydroxpropyl methacrylate, and ethylene glycol dimethacrylate (Lazarov 2007). Aluminum test chambers should be avoided when testing cyanoacrylates, because they may contain catalysts for the polymerization process. The vehicle acetone further enhances the tendency to polymerize spontaneously (Bruze et al. 1995). Due to its excellent adhesive properties with various materials, cyanoacrylate glues may occur in many different occupations. When sticking something to glass, ceramics, or metal surfaces, 2-hydroxy ethyl methacrylate (2-HEMA), 2-hydroxy propyl methacrylate (2-HPMA), and (tri)ethylene glycol dimethacrylate (TEGDMA or EGDMA) are common ingredients of industrial adhesives (Kanerva et al. 1995). Methacrylates and cyanoacrylates do not cross-react.

Some acrylates have higher potential of sensitization than others. In workers with contact to a glue containing different acrylates, some substances like tri- and diethylene glycol diacrylate showed a high number of positive tests, while other ingredients like isobornyl acrylate did not obtain any reaction in patch testing (Kiec-Swierczynska et al. 2005).

For patch-test purposes, a test screening with several acrylates should therefore be carried out when suspecting acrylate allergy. Chemotechnique has the most diverse patch-test tray on methacrylates; however, other companies also provide a suitable screening composition. It was as early as 1975 when Jordan recommended that MMA alone is not a good screening for acrylate allergy (Jordan 1975). Aalto-Korte et al. (2008) recommended 2-hydroxyethyl methacrylate and ethylene glycol dimethacrylate as screeners for occupational contact allergy to anaerobic sealants and acrylic glues. Accelerators, inhibitors, and catalysts, which may be added to the acrylates, can also sensitize.

6 Colophony

Colophony rosin, a naturally occurring rosin from trees, is another natural base of glues and adhesives. Abietic acid, also available as patch-test allergen, and its oxidative derivatives are the causative

allergens (Guin 1995). It ranks high on hit lists of contact allergens because of widespread exposure reaching from occupational use in wood, paper, paint, electronics, metal, and cosmetics industries to household contacts to natural exposure (Sadhra et al. 1997). Some tackifiers for heel and toe stiffeners in shoes are based on colophony and may therefore be relevant allergens in shoe manufacturers (Freeman 1997). Contact dermatitis due to colophony has also been reported for musicians (Lombardi et al. 2003; Gambichler et al. 2004). This is of a special difficulty because the contact of the allergen to the skin cannot be avoided, for example, by using gloves, and the affected persons cannot change their work place/conditions easily.

Medical use of rosin derivatives on tapes, bandages, surgical and dental dressings, wart paints (Lachapelle and Leroy 1990), and hydrocolloid dressings has been reported (Sasseville et al. 1997). For several hydrocolloid dressings, the allergen is a modified ester of colophony, the pentaerythrite ester (Hausen and Kulenkamp 1998). Most patients with a sensitization to colophony report intolerance of brown-colored tapes. However, no report has been found of occupational contact dermatitis to medical colophony resin adhesives.

7 Others

A very special kind of biological adhesives are the so-called fibrin tissue glues. They are two-component systems consisting of fibrinogen and factor XIII in one syringe and thrombin in the second syringe. By adding thrombin to the fibrinogen/factor-XIII mixture, there is a coagulation reaction. Tissue adhesives are used to recombine skin and organ cuts (liver, spleen, etc.), to seal wounds in surgically opened body cavities or vascular prostheses and to stop bleeding. There has been no report of either occupational contact dermatitis or immediate-type contact reactions to this product because skin contact to the medical person is always prevented by gloves worn for reasons of hygiene. Allergic reactions of the recipients are not the focus of this chapter, but it may be mentioned that moderate to severe anaphylactic reactions may appear after repeated instillations of fibrin sealants containing bovine aprotinin. Different surgical procedures in the dental, cardial, vascular, pulmonary, and other fields are described for this problem (Ockenfels et al. 1995; Wuethrich et al. 1996; Shirai et al. 2005; Kober et al. 2008). Aprotinin-specific IgG and IgE can be found in the affected recipients.

8 Patch Testing with Adhesives and Glues

The most important part of allergological diagnosis is to think about possible exposure to glues and adhesives. Optimally, one should only consider patch tests with possible allergenic glue ingredients when the chemical composition of the glue or adhesive is known. This can be determined by asking the manufacturer or, to a lesser extent, by looking at the material data safety sheets. In daily practice, however, this very often fails. Therefore, a glue-screening series makes good practical sense. Table 2 includes information from two major European patch-test provider companies, namely, Chemotechnique and SmartPractice Europe ALG. Their adhesive and glue series were combined in order to get an overview about available glue contact allergens. That not all substances in these trays are glue allergens is worth mentioning; many are plastic allergens such as phthalates, accelerators and inhibitors, or UV adsorbers. To make it easier, the real glue allergens listed in the table are marked in boldface. Screening with the chemical subgroups after getting information about the basic glue composition, for example, epoxy, acrylic, and formaldehyde resin, is proposed.

The plastic and glue series as provided by several suppliers is a mixture of glue and plastic ingredients as well as substances involved in the synthesis of these products. Substances relevant for glues and adhesives are marked in bold. Many of the abovementioned substances are provided by all of the companies, but the ordering number is not mentioned. That is because the substance is not included in the glue and plastic series of this company. As an exception to this rule,

Table 2 Plastics and glue series as recommended by several distributors

	Miscellaneous	Chemotechnique	SmartPractice Europe ALG (allergEAZE)
1	**Abitol**	A-002	D 0915
2	**Abietic acid**	A-001	D 2382
3	**Benzoyl peroxide**	B-007	D 0201
4	Turpentine oil		D 2322
5	*o*-Cresyl glycidyl ether		D 0917
6	**2,6-Ditert-butyl-4-cresol (BHT)**	D-006	D 0110
7	**2-tert-Butyl-4-methoxyphenol (BHA)**	B-022	D 0111
8	**Diphenyl thiourea**	D-025	D 1021
9	**Cyclohexanone resin**	C-027	
10	**2-*n*-Octyl-4-isothiazolin-3-one**	O 004	D 2427
11	*N*, *N*-Dimethyl-*p*-toluidine	D-016	D 0963
Formaldehyde resins			
1	**Toluene-sulfonamide-formaldehyde resin**	T-010	D 0908
2	***p*-tert-Butylphenol-formaldehyde resin**	B-024	D 0030
3	**Resorcinol-formaldehyde resin**	P-005	
4	**Para-tertiary butylphenol**	B-023	D 0920
5	**2-Monomethylol phenol**	M-015	
Epoxies			
1	**Triethylenetetramine**	T-019	D 0905
3	**4-4′-Diaminodiphenyl methane**	D-001	D 0906
4	**Diethylenetriamine**	D-010	
6	**Isophoronediamine**	I-006	
7	**Hexamethylenetetramine**	H-003	D 2318
8	Cresylglycidylether		D 0917
9	**Epichlorhydrin, epoxidharz**	E-002	D 0021
10	**Triglycidyl isocyanurate**	T-028	
11	**Bisphenol A**	B-013	D 0965
Plasticizer			
1	Diethylphthalate		
2	Di-*n*-butylphthalate	D007	D 0903
3	Tricresyl phosphate	T-015	D 2511
4	Triphenyl phosphate	T-022	
5	Dimethylphthalate		D 0954
Plastic stabilizers			
1	2-Phenylindole	P-007	
Plastic inhibitors			
1	Azodiisobutyrodinitrile	A-018	
2	Hydroquinone	H-007	D 0800
3	4-tert-Butylcatechol (PTBC)	B-030B	D 2810
UV adsorbers in plastics			
1	2(2-Hydroxy-5-methylphenyl)benzotriazole	H-016	
2	Resorcinol monobenzoate		
3	Phenyl salicylate		
Isocyanates			
1	Diphenylmethane-4,4-diisocyanate	D-023B	
2	Phenylisocyanate		
3	Toluenediisocyanate		

(*continued*)

Table 2 (continued)

	Miscellaneous	Chemotechnique	SmartPractice Europe ALG (allergEAZE)
Acrylates			
1	**(2-Hydroxyethyl) methacrylate (2-HEMA)**	H-010	D 2477
2	**(2-Hydroxypropyl) methacrylate (2-HPMA)**	H-018	
3	**2-Hydroxyethylacrylate**	H-009	
4	**1,6-Hexanediol diacrylate**	H-004	
5	Tetrahydrofurfuryl methacrylate	T-027	
6	**Tetraethyleneglycol dimethacrylate**	T-029	
7	*N*, *N*-Dimethylaminoethyl methacrylate	D-045	
8	**Methyl methacrylate**	M-013	D 1800
9	**Ethyleneglycol dimethacrylate**	E-007	D 1850
10	**Triethyleneglycol dimethacrylate**	T-018	D 1851
11	**Triethyleneglycol diacrylate**	T-017	
12	**BIS-GMA**	H-013	D 1852
13	**BIS-MA**	M-007	
14	**Butylacrylate**	B-018	
15	***n*-Butyl methacrylate**	B-021	
16	**2-Ethylhexyl acrylate**	E-009	
17	**Ethyl acrylate**	E-004	
18	**Ethyl methacrylate**	E-012	
19	Diurethane dimethacrylate		
20	Urethandimethacrylat	U-004	D 2475

Chemotechnique methacrylates have been included in this table because, in addition to the glue and adhesive series, they provide an extensive list of acrylic adhesives and acrylics for artificial nails which are therefore considered to be adhesives/glues.

Under special circumstances it is also recommended testing the glue itself. When the complete composition is known and the single allergenic ingredients are available, testing the native glue is avoidable. It is warned against testing unknown epoxy resin-based glues because of possible active sensitization. Active sensitization is also well known for acrylics and has been reported even after a single exposure. However, by leaving the glue exposed to air and letting it dry on the patch-test chamber, the risk of strong patch-test reactions is minimized.

The easiest way to patch test a patient's own products is using tapes and medical self-adhesive dressings. Simply cut a small piece and stick it to the skin for 24–48 h. For further information, the other chapters in this book that deal with acrylates and epoxy resins are referred to (see ▶ Chaps. 64, "Coatings," ▶ 56, "Other Plastics," ▶ 207, "Polyvinyl Resins," and ▶ 52, "Epoxy Resins").

9 Other Tests

Since some of the mentioned allergens do not only cause type VI sensitizations with subsequent contact dermatitis but also allergic asthma, it seemed useful to find other methods than patch testing. Elms et al. (2005) tested serum samples for specific IgE antibodies against colophony which could be detected in colophony-exposed workers with symptoms of occupational asthma. The test is based on the production of a protein extract by mono-mac-6 cells after in vitro challenge with colophony extract.

Another method to determine exposure toward allergens is measuring components of allergens in body fluids. Jones et al. (2001) described the analysis of dehydroabietic acid in urine as a biomarker of colophony exposure. There was a

detectable correlation of the levels of dehydroabietic acid in workers of a soldering factory to their colophony exposure.

In terms of contact dermatitis, these tests are of course of lower interest but can give further information about the correlation of the present skin disease and the underlying workplace situation.

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