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Patch-test Reactions to Plastic and Glue Allergens

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Plastics and glues are common causes of occupational dermatoses, but only few reports have dealt with patch-test reactions caused by plastic and glue allergens. Patients exposed to plastics and remitted to an occupational dermatology clinic, were patch-tested with 50 – 53 plastic and glue allergens during a 6-year period. Conventional patch-test techniques were used. The most common causes of allergic patch-test reactions in 360 patients were novolac epoxy resin (5.1%), phenol formaldehyde resin (3.1%), 4-tert-butylcatechol (2.6%), phenyl glycidyl ether (2.6%), diaminodiphenyl methane (2.2%), benzoyl peroxide (2.2%), hexamethylene tetramine (2.0%) and o-cresyl glycidyl ether (1.6%). The allergens that most often elicited irritant patch-test reactions were: benzoyl peroxide (9.5%), abitol alcohol (3.6%), hydroquinone (3.1%), acid-catalyzed phenol formaldehyde resin (2.5%) and toluene diisocyanate (1.9%). Twenty-six out of 53 chemicals caused no allergic reaction during the 6-year period. Plastic allergens in the standard series provoked allergic reactions with formaldehyde (5.8%), diglycidyl ether of bisphenol A (3.2%), 4-tert-butylphenol-formaldehyde-resin (1.1%), toluene sulphonamide formaldehyde-resin (1.1%) and triethylenglycol diacrylate (0.4%). Although half of the plastic chemicals gave no allergic patch-test reactions during a 6-year period, with unusual allergens this low yield needs to be accepted, because otherwise rare allergies will not be detected. Also a negative reaction has diagnostic value.

Key words: acrylate; allergic; benzoyl peroxide; contact dermatitis; diaminodiphenyl methane; epidemiology; epoxy resin; irritant; phenol formaldehyde resin; phenyl glycidyl ether; 4-tert-butylcatechol.

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Plastics and glues are common causes of occupational dermatoses (1 – 6), but only few reports have disclosed data on patch-test reactions caused by plastic and glue allergens (7 – 9). We have compiled 6-year statistics on patch-testing with plastic and glue allergens.

MATERIALS AND METHODS

Most of the patients at our clinic are remitted because of a suspected occupational skin disease (1, 3). They are usually patch-tested with a modified European standard series, other relevant series related to their work or hobbies and their own substances. Patients with exposure to plastics and glues are often patch-tested with our plastics and glues series ("plastics and glues series 1", compounds 1 – 30, and/or "plastics and glues series 2", compounds 31 – 50; Table I). During part of the time 2 additional allergens were patch-tested (substances 30a and 30b in Table I). We have compiled our 6-year patch-test results with these 50 – 53 allergens (Table I). For comparison, we have analysed our 6-year results of plastic allergens in the European standard series (Table II).

Patch-testing was performed as previously described (1, 3, 10) with 2 days' occlusion and 3 readings (usually on days 2, 3 and 4 – 6). Allergic reactions were scored according to ICDRG recommendations. 1+ (palpable erythema, papules), 2+ (oedematous, vesicles) and 3+ (bullous) reactions were considered allergic. Irritant reactions were also recorded. Reactions scored as doubtful (?+) or irritant (IR) were classified as irritant. The patch-test substances were from Chemotechnique Diagnostics AB (Malmö, Sweden), Trolab, Hermal Chemie (Reinbeck/Hamburg, Germany); Epikon Oy (Helsinki, Finland) or were manufactured by ourselves (Table I).

RESULTS

The number of patients studied, allergic and irritant patch-test reactions, source of patch-test substances, and the common use of the plastics and glues are summarized in Table I. The 10 most common causes of allergic patch-test reactions (in rank order) were novolac epoxy resin (5.1%), phenol formaldehyde resin (3.1%), 4-tert-butylcatechol (2.6%), phenyl glycidyl ether (2.6%), diaminodiphenyl methane (2.2%), benzoyl peroxide (2.2%), hexamethylene tetramine (2.0%) and o-cresyl glycidyl ether (1.6%). Allergens that most often elicited an irritant reaction were benzoyl peroxide (9.5%), abitol alcohol (3.6%), hydroquinone (3.1%), acid-catalyzed phenol formaldehyde resin (2.5%) and toluenediisocyanate (TDI; 1.9%). Twenty-six out of 53 chemicals caused no allergic reaction during a 3-year period (Table I).

Patch-test reactions with 5 plastic allergens in the standard series are shown in Table II: formaldehyde provoked an allergic reaction in 5.8% of the patients, followed by diglycidyl ether of bisphenol A epoxy resin (3.2%), 4-tert-butylphenol-formaldehyde-resin (1.1%), toluene sulfonamide formaldehyde resin (1.1%) and triethyleneglycol diacrylate (0.4%).

DISCUSSION

In Finland the most common occupational plastic and glue allergens are the epoxy resins (1, 3, 5, 10) and acrylic resins (11, 12), and we have separate patch-test series for these allergens. For example, dental personnel who have the greatest incidence of allergy to acrylics are usually not patch-tested with the plastics and glues series. Triethyleneglycol diacrylate (TREGDA) has been included in our standard series. Although it gave an allergic reaction in only 6/1425 (0.4%) of the patients tested, in some cases these allergic patch-test reactions have given a clue to the cause of the patient's occupational allergic skin disease (13). Allergic patch-test reaction frequencies with the (meth)acrylates are much higher than with plastic and glue allergens: 2-hydroxyethyl methacrylate (2-HEMA), the common allergen in dental acrylics, provoked 11.4% allergic patch-test reactions in a 10-year study (12).

We have earlier published our 3-year results on plastic and glue allergens in a short communication (9). It showed the same trend as in this extended study. The most common plastic

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Table I. *Allergic and irritant patch-test reactions to a plastic and glue series during 1991–96. Allergens 1–30 = "plastics and glues series 1" and allergens 31–50 = "plastics and glues series 2"*

Number	Compound	Use/exposure	Patch-test concentration; % (w/w)	*	1991–96			1991–93	1994–96	1991–96	1991–96	1991–96	1998–
					Allergic/ tested	Allergic (%)	Rank order (top ten)	Allergic/ tested	Allergic / tested	Irritant/ tested	Irritant (%) (frequency > 1%)	Irritant/ Rank order	Allergen included in the plastics and glues series since 1998**
1	Hydroquinone	Inhibitor in acrylic monomers; anti-oxidant in animal feed; photographic reducer and developer	1.0	C	0/357	–		0/180	0/177	11/357	3.1	3	yes
2	Dibutyl phthalate	Plasticizer; insect repellent; emollient in aerosol antiperspirants	5.0	C	0/357	–		0/180	0/177	2/357			yes
3	Phenyl salicylate	UV-light absorber in plastics, suntan oils, and creams	1.0	C	0/358	–		0/180	0/178	1/358			yes
4	Dioctyl phthalate	Plasticizer	2.0	C	0/357	–		0/180	0/177	2/357			yes
5	2,6-Di-tert-butyl-4-cresol (BHT)	Antioxidant	2.0	C	0/358	–		0/180	0/178	2/358			yes
6	2(2-Hydroxy-5-methyl-phenyl) benzotriazol	UV-absorber	1.0	C	0/357	–		0/180	0/177	2/357			yes
7	Benzoylperoxide	Initiator in the polymerization of plastics	1.0	C	8/357	2.2	7.	2/180	6/177	34/357	9.5	1	yes
8	1,4-Butanediol dimethacryl-ate (BUDMA)	Cross-linking methacrylic monomer for use in dental composite materials, sealants, prostheses, etc.	2.0	C	1/355	0.3		1/178	0/177	0/355			no
9	Azodiisobutyrodi-nitrile	Inhibitor and foaming agent in plastic and elastomer materials	1.0	C	0/358	–		0/180	0/178	2/358			yes
10	Bisphenol A	Raw material in the production of epoxy and acrylic resins	1.0	C	1/356	0.3		0/180	1/176	1/356			no
11	Tricresyl phosphate	Plasticizer in vinyl plastics, spectacle frames	5.0	C	0/357	-		0/180	0/177	6/357	1.7	6	yes
12	Phenol formaldehyde resin	Resin used in binders, adhesives, laminates, impregnation products, surface coatings, etc.	1.0	C	11/357	3.1	2.	3/180	8/177	5/357	1.4	9	yes
13	Triphenyl phosphate	Plasticizer in plastics, lacquers, varnishes, etc.	5.0	C	1/358	0.3		0/181	1/177	3/358			yes
14	Novolac epoxy resin	Epoxy resin; does not necessarily cross-react with standard epoxy resin	1.0	F	9/178	5.1	1.	ND	9/178	2/178			no
15	Resorcinol monobenzoate	UV-light absorber in outdoor plastics	1.0	C	2/357	0.6		2/180	0/177	0/357			yes
16	2-Phenylindole	Stabilizer in PVC-plastic products	2.0	C	0/357	–		0/179	0/178	2/357			yes
17	2-tert-Butyl-4-methoxy-phenol (BHA)	Antioxidant	2.0	C	0/358	–		0/180	0/178	3/358			yes
18	Triethylenetetramine (TETA)	Epoxy curing agent	0.5 aq	C	0/356	–		0/180	0/176	3/356			no
19	Diethylenetriamine (DETA)	Epoxy curing agent	1.0	C	1/356	0.3		0/180	1/176	2/356			no
20	Diaminodiphenyl-methane (MDA)	Epoxy curing agent	0.5	C	8/356	2.2	7.	5/180	3/176	3/356			yes
21	Abitol alcohol	Plasticizer; also in adhesives, mascaras, inks, sealants, etc.	10.0	C	0/359	–		0/181	0/178	13/359	3.6	2	yes
22	4-tert-Butylphenol	Antioxidant in plastics, adhesives, etc.; intermediate in the production of lacquer and varnish resins	1.0	C	0/359	–		0/181	0/178	3/359			yes
23	Toluene diisocyanate (TDI)	Production of polyurethane foams, elastomers, adhesives, printing plates, etc.	1.5/2.0	O/C	3/360	0.8		2/181	1/179	7/360	1.9	5	no
24	Diphenylmethane-4, 4-diisocyanate (MDI)	Production of polyurethane lacquers, foams plastics, rubber, and glues	1.5/2.0	F/C	3/360	0.8		3/181	0/179	2/360			no

Number	Compound	Use/exposure	Patch-test concentration; % (w/w)	*	1991–96			1991–93	1994–96	1991–96	1991–96	1991–96	1998–
					Allergic/ tested	Allergic (%)	Rank order (top ten)	Allergic/ tested	Allergic / tested	Irritant/ tested	Irritant (%) (frequency > 1%)	Irritant/ Rank order	Allergen included in the plastics and glues series since 1998**
25	2-Monomethylol phenol	Intermediate in the production of phenol formaldehyde resins	1.0	C	3/359	0.8		0/180	3/179	2/359			yes
26	Methyl methacrylate (MMA)	Methacrylic monomer in plastics for dentures, bone cement, artificial nails, hearing aids, etc.	2.0	C	2/356	0.6		2/180	0/176	5/356	1.4	9	no
27	Phenol formaldehyde resin, acid catalyzed	Resin used in binders, adhesives, laminates, impregnation products, surface coatings, etc.	5.0	T	9/360	2.5	5.	3/181	6/179	9/360	2.5	4	no
28	Hexamethylene-tetramine	Epoxy curing agent; rubber accelerator	2.0	C	7/357	2.0	9.	4/180	3/177	2/357			no
29	Diphenyl thiourea	Rubber chemical	1.0	T	1/360	0.3		0/181	1/179	1/360			yes
30	Phenol formaldehyde resin, alkali catalyzed	Resin used in binders, adhesives, laminates, impregnation products, surface coatings, etc.	5.0	T	0/360	–		0/181	0/179	3/360			yes
30a	Diphenylmethane-4, 4-diisocyanate (MDI)	Production of polyurethane lacquers, foams, plastics, rubber, and glues	2.0	F	0/179	–		ND	0/179	0/179			no
30b	Toluene sulfonamide formaldehyde resin	Modifier and adhesion promoter for film-forming resins, e.g., nail lacquers, acrylics	10.0	C	1/269	0.4		1/181	0/88	2/269			no
31	Urea formaldehyde resin	Resin in formaldehyde resin plastics	10.0	T	4/308	1.3		1/150	3/158	4/308	1.3	11	yes
32	Melamine formaldehyde resin	Resin in formaldehyde resin plastics	10.0	T	3/309	1.0		0/150	3/159	3/309			yes
33	alpha-Pinene	Principal constituent of oil of turpentine	15.0	T	0/310	–		0/151	0/159	1/310			yes
34	4-tert-Butylcatechol	Prevents polymerization of polyester resin, butadiene and PVCs; stabilizing agent in plastics, e.g., monomeric styrene, butadiene, isoprene	1.0	T	8/308	2.6	3.	3/150	5/158	5/308	1.6	7	yes (0.5% and 0.25%)
35	Diethyl phthalate	Plasticizer	5.0	T	0/309	–		0/150	0/159	2/309			no
36	Diisodecyl phthalate	Plasticizer	5.0	T	0/310	–		0/151	0/159	2/310			no
37	Dimethyl phthalate (DMP)	Plasticizer	5.0	T	0/310	–		10/151	0/159	3/310			yes
38	Di-2-ethylhexyl phthalate (DEHP)	Plasticizer	5.0	T	0/310	–		0/151	0/159	3/310			yes
39	Isophoronediamine (IDPA)	Epoxy curing agent	0.5	T	0/311	–		0/152	0/159	1/311			no
40	o-Cresyl glycidyl ether	Reactive diluent in epoxy resin systems	0.25	T	5/311	1.6	10.	3/153	2/158	5/311	1.6	7	no
41	*n*-Butyl glycidyl ether	Reactive diluent in epoxy resin systems	0.25	T	2/310	0.6		1/152	1/158	1/310			no
42	Phenyl glycidyl ether	Reactive diluent in epoxy resin systems	0.25	T	8/309	2.6	3.	5/151	3/158	2/309			no
43	Phenyl isocyanate	Production of polyurethanes	0.1	T	0/310	–		0/151	0/159	3/310			no
44	Triphenylmethane triisocyanate	Production of polyurethanes	0.1	T	0/310	–		0/151	0/159	2/310			no
45	N, N-Dimethyl-4-toluidine	Amine accelerator for the polymerization of e.g. dental acrylics	5.0	C	0/309	–		0/150	0/159	1/309			yes
46	Epichlorohydrin	Raw material in the production of epoxy and acrylic resins	0.1	T	0/308	–		0/151	0/157	2/308			no
47	Abietic acid	Major component of resin used in adhesive tapes, glues, inks, sealants, cosmetics, dental impression materials	10.0	C	4/307	1.3		2/149	2/158	3/307			yes
48	Acrylonitrile	In the production of acrylonitrile-butadiene-styrene and styrene--acrylonitrile plastics	0.1	E	0/308	–		0/149	0/159	1/308			no
49	Cyanoacrylate	Adhesives for plastics, rubber, glass, metals; "super glues"	2.0	E	1/306	0.3		1/149	0/157	2/306			no
50	Phenol formaldehyde resin	Resin used in binders, adhesives, laminates, impregnation products, surface coatings, etc.	5.0	F	8/308	2.6	5.	1/149	7/159	5/308	1.6	7	yes

Test substances mixed in petrolatum (pet.) except when indicated as aq: mixed in aqua.
*Source of allergen: C, Chemotechnique Diagnostics AB; T, Trolab Hermal Chemie; F, Finnish Institute of Occupational Health; E, Epikon Oy.
**Cyclohexanone resin (C) has been added to the plastic and glues series.

Table II. *Patch-test results of plastic and glues allergens in the standard series*

Number	Compounds in standard series	Use/exposure	Patch-test concentration: % (w/w)	*	Allergic/ tested	Allergic (%)
1	Diglycidyl ether bisphenol A (DGEBA)	Epoxy resin for use in surface coatings, adhesives, electrical insulation, etc.	1.0	C	46/1416	3.2
2	Triethyleneglycol diacrylate (TREGDA)	Cross-linking acrylate monomer for use in coatings, adhesives, and in printing plates of photoprepolymer type	0.1	C	6/1425	0.4
3	Formaldehyde	Antimicrobial; wide use in production of plastics	1.0	C	82/1414	5.8
4	4-tert-Butylphenolform-aldehyde-resin (PTBP)	Resin used in adhesives for shoes and watch straps; do-it-yourself glues, plywood, insulation, automobiles, motor oils, etc.	1.0	C	16/1422	1.1
5	Toluene sulphonamide formaldehyde resin	Modifier and adhesion promoter for film forming resins, e.g. nail lacquers, acrylics	10.0	C	6/525	1.1

* Source of allergen.
C = Chemotechnique Diagnostics Ab.

allergens are epoxy resins, including standard epoxy resin, novolac epoxy resin, the epoxy reactive diluents phenyl glycidyl ether, o-cresyl glycidyl ether (10), phenol formaldehyde resin, 4-tert-butylcatechol (14) and diaminodiphenyl methane, benzoyl peroxide and hexamethylene tetramine. Formaldehyde is used in the plastic industry, but it has also many other applications, and is a more common cause of allergic patch-test reactions than the above-mentioned substances.

All 9 of our patients who had an allergic patch-test reaction from novolac epoxy resin (Table I, substance 14) reacted to DGEBA-ER. When analysed novolac epoxy resin contained traces of DGEBA-ER, which explains the reactions. Recently, Erikstam & Bruze (15) reported more patch-test reactions with bisphenol F (i.e. novolac epoxy resin) than with DGEBA-ER, but the purity and the exact composition of bisphenol F was not revealed.

Tarvainen (7) reported on allergic patch-test reactions at a university dermatology clinic in which most of the patients are not investigated because of a suspected occupational skin disease. The most frequent allergens in her study were: *p*-tert-butylphenol formaldehyde resin (2.6%), abietic acid (2.3%), phenyl glycidyl ether (1.2%), and diaminodiphenyl methane (0.9%). Fourteen out of 32 allergens caused no allergic reaction in her 839 patients.

Toluene sulphonamide formaldehyde resin, which is a modifier and adhesion promoter for film forming resins, and is used in most nail lacquers (16 – 18) gave only 1.1% allergic reactions when tested in our standard series (Table II) and 0.4% in our plastic and glues series, whereas in non-occupational clinics much higher allergy frequencies have been reported, e.g. 6.6% in an Italian study (17). This reflects the great variations in patient materials in occupational vs. non-occupational dermatology clinics.

Holness & Nethercott (8) reported results from a patch-test clinic having mainly patients suspected of an occupational skin disease. Their most common allergens were phenyl glycidyl ether (6.1%), ethylenediamine (3.1%), melamine formaldehyde resin (3.1%) and triethylene tetramine (2.5%). Twenty allergens caused at least 1 allergic patch-test reaction, whereas 26 allergens elicited no allergic reaction. On the other hand, 30 individuals, i.e. 12.8% of those tested, had at least 1 positive response.

In earlier (7 – 9), and the present report the relevance (past or present) of the patch-test reactions has not been determined. Based on the history and clinical picture, most of our allergic patch-test reactions were relevant to past or present eczema (related to past or present exposure). Some of the allergic reactions to benzoyl peroxide were difficult to trace, and may represent irritant reactions. Unfortunately, we do not computerize data on "past or present relevance" of the patch-test reactions.

Active sensitization is a complication of patch-testing (19). The compound 4-tert-butylcatechol 1% pet caused several cases of active sensitization as recently reported in detail (14). We have recommended to use a lower patch-test concentration with this chemical, and are currently patch-testing 4-tert-butylcatechol at 0.5% and 0.25% pet (14, Table I). In the present study 1 further patient was sensitized to diglycidyl ether of bisphenol A epoxy resin in the standard series, 1 patient to diaminodiphenylmethane, and 1 to both phenyl-glycidyl ether and diaminodiphenylmethane in the plastic and glues series (20). Patch-testing is usually safe, and as stated by Cronin (21), active sensitization is a complication of patch-testing, but not a hazard, and it should not be used as an excuse for not doing this investigation.

We reported earlier that the "cost/benefit" ratio, i.e. the percentage of allergic reactions to patch-test allergens in the plastics and glues series is not very good (9). In the present study 26 out of 53 chemicals gave no allergic patch-test reactions during

a 6-year period. However, with rare allergens this low yield needs to be accepted, because otherwise rare allergies will not be detected. Furthermore, a negative reaction also has diagnostic value. The allergens that gave allergic patch-test reactions in the present study should probably be chosen if a smaller series for plastics and glues needs to be used. On the other hand, it may be necessary to extend even our large series, depending on exposure, because many other plastic and glue chemicals are allergens, e.g. polyfunctional aziridine hardeners (22). In occupational dermatology, it is also important to patch-test with the patients' "own" substances. Based on the present findings we have revised (Table I) our plastic and glues series and replaced some of the substances that have not given allergic patch-test reactions; partly the allergens have been included into other series, e.g. epoxy resin compounds.

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