

Screening for acrylate/methacrylate allergy in the baseline series: our experience in Sweden and Singapore

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Background: No studies to specifically determine the prevalence of contact allergy to acrylates/methacrylates in patch tested populations have been published.

Objectives: To determine the prevalence of acrylate/methacrylate allergy in all patients tested to the baseline patch test series.

Methods: Five acrylate/methacrylate allergens (2-hydroxyethyl methacrylate, methyl methacrylate, ethylene glycol dimethacrylate, triethylene glycol diacrylate, and 2-hydroxypropyl acrylate) were included in the baseline series for at least 2 years in Malmö and Singapore.

Results: Thirty-eight patients in total had reacted to acrylate/methacrylate allergens in the baseline series during the study period in both populations. In Malmö, there were 26 (1.4%) patients with positive patch tests to acrylate/methacrylate allergens, 14 of whom had relevant reactions. In Singapore, there were 12 (1.0%) patients with positive patch tests to acrylate/methacrylate allergens, but only 1 had relevant reactions. If we had not added acrylate/methacrylate allergens to the baseline series, we would not have patch tested 13/26 (50%) of the positive reactors in Malmö and 11/12 (92%) of the positive reactors in Singapore. The overall proportion of missed positive reactors would have been 24/38 (63%).

Conclusions: The prevalence of acrylate/methacrylate allergy in our patch tested dermatitis populations is 1.4% in Malmö and 1.0% in Singapore.

Key words: allergic contact dermatitis; acrylates; baseline series; patch test; screening. © Blackwell Munksgaard, 2008.

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Although numerous case reports and studies on allergic contact dermatitis to acrylate/methacrylate allergens have been published, there have not been any studies specifically to determine the prevalence of contact allergy to acrylates/methacrylates in entire populations of patients patch tested in a dermatology centre.

However, the prevalence of contact allergy to methyl methacrylate (MMA) and ethyl acrylate has been mentioned in studies reporting patch test results from centres in North America (1, 2). In those studies, the prevalence of contact allergy to

MMA had been reported to be between 0.9% and 1.2%, while that of ethyl acrylate had been reported to be between 0.9% and 1.8%.

Materials and Methods

This is a study of patients patch tested to the baseline patch test series in two dermatology centres – the Department of Occupational and Environmental Dermatology, Malmö University Hospital, Malmö, Sweden and National Skin Centre, Singapore. Five acrylate/methacrylate allergens, obtained from

Chemotechnique Diagnostics (Vellinge, Sweden), were included in the baseline series of both centres. We had ensured that the allergens used in both centres were from the same batches from the manufacturer. These allergens were applied to all patients for whom the baseline series had been ordered over virtually the same period of at least 2 years in both centres.

Patch testing

The allergens were applied onto small (8 mm) Finn Chambers[®] (Epitest Ltd Oy, Tuusula, Finland) on Scanpor[®] (Norgesplaster A/S, Vennesla, Norway). The tests were applied to the upper part of the back and left for 2 D. Tests were read on D3 or D4 and D7 by a dermatologist, and the reactions were scored according to International Contact Dermatitis Research Group criteria.

Allergens

The acrylate/methacrylate allergens added to our baseline series were 2-hydroxyethyl methacrylate (2-HEMA) 2.0% petrolatum (pet.) w/w, MMA 2.0% pet. w/w, ethylene glycol dimethacrylate (EGDMA) 2% pet. w/w, triethylene glycol diacrylate (TREGDA) 0.1% pet. w/w, and 2-hydroxypropyl acrylate (2-HPA) 0.1% pet. w/w. The chemical structures of these allergens are shown in Fig. 1.

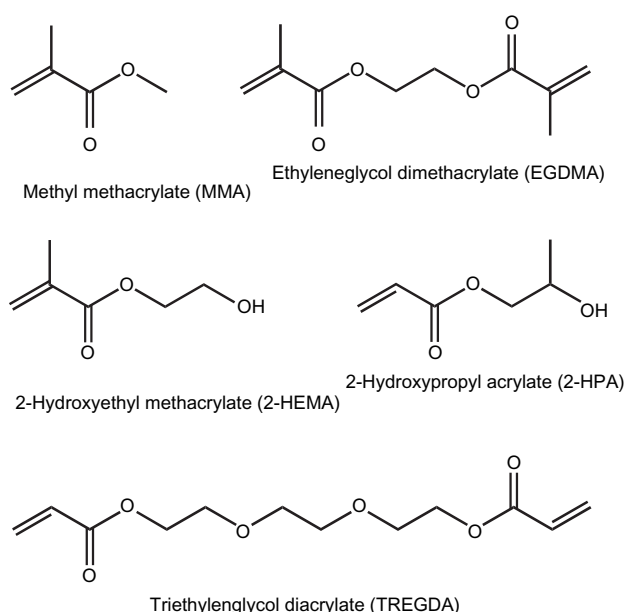


Fig. 1. Chemical structures of the five acrylate/methacrylate allergens included in the baseline patch test series in Malmö and Singapore.

Patients

In Malmö, 1609 (632 male and 977 female) patients (mean age 44 years, range 12–94 years) were tested with the baseline series from 21 February 2005 to 30 June 2007. Where necessary, some of these patients were also tested with supplementary series containing acrylates/methacrylates. These were the dental personnel, (industrial) acrylate, and nail acrylics series.

In Singapore, 1181 (547 male and 634 female) patients (mean age 39 years, range 4–87 years) were tested with the baseline series from 1 July 2005 to 30 June 2007. For some patients, acrylate/methacrylate allergens were also tested in separate acrylate and dental series when deemed necessary.

Statistics

The statistical analyses were conducted using SPSS 12.0.1 for Windows (Chicago, IL, USA). We regarded $P < 0.05$ as statistically significant. The difference in the prevalence of contact allergy and relevance of positive reactions between the Malmö and the Singapore patients was tested using the two-tailed Fisher's exact test.

To compare differences in terms of strengths of reactions, each patient was subclassified according to what his/her strongest positive reaction for the three main allergens (2-HEMA, TREGDA, and EGDMA; i.e. MMA and 2-HPA were omitted because there were no reactions to them in the Singapore material) was. Only patients who had undergone both readings were included in this analysis. Differences between patients whose strongest reactions were + versus ++ versus +++ were compared using the Mann–Whitney *U*-test.

Results

The patch test results of the patients with positive reactions to acrylate/methacrylate allergens in our study populations are shown in Table 1.

In Malmö, there were 26 (1.6%) of 1609 patients (mean age 46 years, range 23–81 years) with positive patch tests to acrylate/methacrylate allergens in the baseline series. There were 9 (1.4%) of 632 males (mean age 45 years, range 24–72 years) and 17 (1.7%) of 977 females (mean age 46 years, range 23–81 years) in this group. The allergens, in order of frequency, were 2-HEMA > TREGDA > 2-HPA > EGDMA > MMA.

In Singapore, there were 12 (1.0%) of 1181 patients (mean age 38 years, range 19–61 years) with positive patch tests to acrylate/methacrylate

Table 1. Patients with positive reactions to acrylates/methacrylates in the baseline series in Malmö and Singapore*

Patient no.	Sex	Age (years)	Hist	Rel	2-HEMA		MMA		EGDMA		TREGDA		2-HPA	
					D3/D4	D7	D3/D4	D7	D3/D4	D7	D3/D4	D7	D3/D4	D7
M1	F	81	N	N	—	?	?	+	—	?	—	—	—	—
M2	F	74	N	N	—	+	—	—	—	?	—	—	—	—
M3	F	74	N	N	—	—	?	—	+++	+++	?	—	—	—
M4	M	72	N	N	—	—	—	+	—	—	—	—	—	—
M5	M	64	N	N	—	—	—	—	—	—	++	+	—	—
M6	M	62	N	N	+++	+++	—	—	+	+	—	—	—	—
M7	F	60	Y	Y	++	++	—	—	—	++	—	—	—	—
M8	F	55	N	Y	?	++	—	—	—	?	—	?	+	++
M9	F	55	N	N	?	+	—	—	—	—	+	+	—	—
M10	M	49	N	N	++	++	—	—	++	++	—	—	—	—
M11	F	48	Y	Y	++	+	—	—	+	+	+	+	+	+
M12	F	47	Y	Y	+++	++	—	—	+++	++	—	—	++	+
M13	F	43	Y	Y	+	+	—	—	+	+	—	—	—	—
M14	F	41	Y	Y	—	—	—	—	—	—	+	?	—	—
M15	M	41	Y	Y	—	—	—	—	—	—	+	?	+	?
M16	F	39	N	N	—	+	—	—	—	—	—	—	—	—
M17	M	33	N	N	—	+	—	—	—	—	—	—	—	?
M18	F	33	N	N	—	—	—	—	—	—	—	+	—	—
M19	F	31	Y	Y	++	++	—	—	++	++	—	—	+	+
M20	F	28	Y	Y	—	—	—	—	—	—	+	+	—	—
M21	M	28	Y	Y	—	—	—	—	—	—	+	—	+	—
M22	M	28	N	N	++	+	—	—	—	++	—	—	+	+
M23	F	27	Y	Y	+	+	—	—	+	+	+	—	+	+
M24	F	23	Y	Y	—	—	—	—	—	—	+++	+	+	—
M25	M	24	Y	Y	?	?	—	—	—	—	++	+	++	+
M26	F	23	Y	Y	?	+	—	—	—	—	+++	++	+	+
S27	F	24	N	N	+	—	—	—	—	—	—	—	—	—
S28	F	61	N	N	—	—	—	—	+	—	—	—	—	—
S29	F	23	N	N	—	—	—	—	+	?	—	—	—	—
S30	M	25	N	N	—	—	—	—	—	—	+	—	—	—
S31	F	41	N	N	—	NR	—	NR	—	NR	+	NR	—	NR
S32	M	19	N	N	—	—	—	—	—	—	?	+	—	—
S33	F	48	N	N	—	—	—	—	—	—	?	+	—	—
S34	M	56	N	N	—	—	—	—	—	—	+	—	—	—
S35	M	37	N	N	—	—	—	—	+	—	+	—	—	—
S36	F	27	N	N	+	—	—	—	—	—	—	—	—	—
S37	M	55	N	N	—	—	—	—	+	—	—	—	—	—
S38	F	44	Y	Y	+	NR	—	NR	+	NR	—	NR	—	NR

?, doubtful positive reaction; 2-HEMA, 2-hydroxyethyl methacrylate 2% petrolatum (pet.); 2-HPA, 2-hydroxypropyl acrylate 0.1% pet.; D3/D4, first reading; D7, second reading; EGDMA, ethylene glycol dimethacrylate 2% pet.; Hist, history suggestive of contact allergy to acrylate/methacrylate allergens; M1–M26, patients from Malmö; MMA, methyl methacrylate 2% pet.; N, no; NR, not read; pet., petrolatum; Rel, relevance found for positive reaction(s) to acrylate/methacrylate allergens; S27–S38, patients from Singapore; TREGDA, triethylene glycol diacrylate 0.1% pet.; Y, yes.

*All patch test readings are shown as first reading (D3/D4)/second reading (D7).

allergens in the baseline series. There were 5 (0.9%) of 547 males (mean age 38 years, range 19–56 years) and 7 (1.1%) of 634 females (mean age 38 years, range 23–61 years) in this group. The positive reactions, in order of frequency, were: TREGDA > EGDMA > 2-HEMA. There were no positive reactions to MMA and 2-HPA.

The difference in proportions of positive reactors in the patch test populations between the two centres was not statistically significant ($P = 0.19$) with the two-tailed Fisher's exact test.

Overall, 38 patients in total had reacted to acrylate/methacrylate allergens in the baseline series during the study period in both populations. The overall ranking in number of positive reactions

was 2-HEMA = TREGDA > EGDMA > 2-HPA > MMA.

Among the positive reactors in Malmö, the most common areas of involvement with dermatitis were the hands (18), face (5), and chest (5); while the most common areas of involvement in Singapore were the face (5) and hands (4).

Of the 26 positive reactors in Malmö, 13 gave a history suggestive of contact allergy to acrylate/methacrylate allergens and had been simultaneously tested to the supplementary series. In Singapore, only 1 of the 12 patients had given a history suggestive of contact allergy to acrylate/methacrylate allergens and had been simultaneously tested to the supplementary series. The majority of our

patients with positive reactions had not, prior to patch testing, given a history suggestive of contact allergy to acrylate/methacrylate allergens.

Among the 26 positive reactors in Malmö, all had come for both readings. Of these 26 patients, many had also been tested to and had positive reactions to acrylate/methacrylate allergens in supplementary series: 8 with the nail acrylics series, 6 with the (industrial) acrylate series, and 3 with the dental personnel series. Three of those with the (industrial) acrylate and one with the nail acrylic series had the supplementary series added only after the first reading, where the screening allergens had picked up potential acrylate/methacrylate allergy. Among the 12 positive reactors in Singapore, 10 had come for both readings, while 2 had defaulted the D7 reading. One of these defaulters (patient no. 38) had also been tested to and had positive reactions to acrylate/methacrylate allergens in the acrylate series on D3. Therefore, if we had not added acrylate/methacrylate allergens to the baseline series, we would not have picked up 13 (50%) of the 26 positive reactors in Malmö and 11 (92%) of the 12 positive reactors in Singapore. The overall proportion of missed positive reactors would be 24 of 38 or 63%. However, of these 24 missed positive reactors, only 1 (patient no. 8) had been found to have relevant positive reactions to acrylate/methacrylate allergens. This was a 55-year-old administrator who worked in a warehouse. She had a relevant history of dermatitis around her fingernails from acrylic nails.

Twenty per cent of Singapore patients had not attended for their second readings, including two of the positive reactors. This may mean that the proportion of missed reactors in Singapore may be even higher than the estimated value of 92%.

Discussion

This is the first study where the entire patch tested populations patch tested to the baseline series in any centres have been tested to these five acrylate/methacrylate allergens. The selection of allergens to be included in the baseline series was aided by retrospective data from testing with acrylate/methacrylate allergens from the supplementary series containing these allergens in both centres. In Malmö, the commonest allergens, in order of frequency, had been 2-HEMA > EGDMA > triethylene glycol dimethacrylate (TREGDMA) > TREGDA > MMA (3, 4). In Singapore, these had been EGDMA > 2-HPA > 2-HEMA > TREGDA (3, 4).

In the present study, we had included 2-HEMA and EGDMA into the baseline series because these two allergens had given the highest yield

when testing dental patients in Malmö. MMA had been chosen partially due to historical reasons, as it had been suggested in the international literature as a marker for acrylate allergy (5). TREGDA had been chosen because it is an industrial acrylate that would give us an industrial aspect of this contact allergy. 2-HPA had been chosen because it had been one of the allergens commonly giving positive reactions in Singapore (second only to EGDMA), with a high frequency of simultaneous positive reactions with EGDMA.

There were two patients in each centre (not included in Table 1) who did not react to the acrylate/methacrylate allergens in the baseline series but reacted to other acrylate/methacrylate allergens in the supplementary series. From Malmö, one male patient had reacted to trimethylolpropane triacrylate (TMPTA) and pentaerythritol triacrylate in the (industrial) acrylate series, while another female patient had reacted to ethylcyanoacrylate (ECA) in the nail acrylics series (6). From Singapore, one female dentist had reacted to tetrahydrofurfuryl methacrylate in the dental series, while another female nail technician had reacted to TMPTA, tripropyleneglycol diacrylate, and urethane diacrylate (aliphatic) in the acrylate series. These four patients had not been included in our analysis.

The majority of our patients with positive reactions had not given any history suggestive of contact allergy/allergic contact dermatitis to acrylate/methacrylate allergens. Possible explanations could be inadequate history-taking by the doctors or that patients with a lower degree of acrylate/methacrylate allergy had perceived neither an association between a possible acrylate/methacrylate exposure and dermatitis nor that the contact allergy had manifested as allergic contact dermatitis. A statistical difference in strength of reaction between those with a history of contact allergy and those without would speak for the low reactivity interpretation. However, there was no difference in strength of reaction between those who had given a history suggestive of contact allergy to acrylates/methacrylates and those who had not ($P = 0.14$, Mann–Whitney U -test). The lack of statistical significance may be due to the small number of positive reactors in our population.

Of the 26 positive reactors in Malmö, 14 (54%) had been found to have relevant reactions to acrylate/methacrylate allergens; 6 female nail technicians, 3 female dental workers, 3 men who worked with ultraviolet-cured acrylates, and 2 female acrylic nail users. Of the 12 positive reactors in Singapore, only 1 had been found to have relevant reactions to acrylate/methacrylate allergens. This was the aforementioned 44-year-old female nail

technician (patient no. 38) who had also reacted to acrylate/methacrylate allergens in the acrylate series as well as her own nail acrylic products. The difference in proportions of relevant reactions between the two centres is significant ($P = 0.012$, two-tailed Fisher's exact test). The differences in relevance between the two centres may reflect less rigorous probing or less available time for probing of positive reactors' previous exposure to acrylates by Singapore dermatologists. As discussed earlier, patients with a low degree of acrylate allergy may fail to obtain a manifest allergic contact dermatitis from acrylates when exposed, and the interpretation may be that it is a non-relevant contact allergy. Indeed, there is some support to the low reactivity interpretation as the comparison of strength of reactions between those with relevant reactions compared with those without relevant reactions was close to being statistically significant ($P = 0.07$, Mann–Whitney U -test). Furthermore, although no relevance was found for a large number of the reactions, future relevance may be important. Therefore, patients in whom no relevance was found should still be advised to avoid/minimize contact with acrylate/methacrylate allergens in order to prevent future allergic contact dermatitis. They may be advised to avoid artificial nails as there may be a higher incidence of allergic contact dermatitis if they are exposed. They may also be advised to inform their dentists of their contact allergy even though there may not be a high incidence of stomatitis even if they are exposed. The dentist may then be alerted to use rubber dams when making white plastic fillings in these patients.

In Malmö, one patient (not included in Table 1) who had negative readings to 2-HEMA on D3 and D7 was found to have a positive reaction to this allergen when she was seen again 1 month after the patch test application. We have reported that late-appearing reactions to 2-HEMA and EGDMA beyond D7 may occur at the current internationally recommended patch test concentrations (7). These may not necessarily be due to active sensitization (7).

Again, the importance of a D7 reading should also be highlighted (3, 4, 7). If only one reading had been made (on D3 but not also on D7), we would have missed 40% 2-HEMA-positive, 8% TREGDA-positive, 20% EGDMA-positive, and 100% MMA-positive reactions in the Malmö patients as well as 33% TREGDA-positive reactions in our Singapore patients. Three patients with relevant positive reactions would have been missed (patients 7, 8, and 26). Patient 7 was a 60-year-old administrator in a cosmetic company with relevant history of dermatitis around her nails

from acrylic nails. Patient 26 was a 23-year-old nail technician with current dermatitis on her fingertips, wrists, forearms, and face from nail acrylates.

In the total population of patients patch tested with the baseline series in the two centres during the study period, almost all patients from Malmö had undergone two readings, while 939 of 1181 (80%) patients from Singapore had undergone both readings. When considering only those patients who have had two patch test readings, we can estimate that 13 of 1609 (0.8%) patients from Malmö and 10 of 939 (1.1%) patients from Singapore would have been missed if acrylate/methacrylate allergens had not been screened for in the baseline series. These figures give a reflection of the possible proportions of acrylate-allergic/methacrylate-allergic patients that we have been missing among our patch tested populations.

In our previous retrospective studies (3, 4) in Malmö, we had found that a screening series of seven allergens would have picked up all our past patients (dental, industrial, and nail) with suspected allergy to acrylate/methacrylate allergens. These allergens were 2-HEMA, TREGDA, 2-hydroxypropyl methacrylate (2-HPMA), 2,2-bis[4-(2-hydroxy-3-methacryloxypropoxy) phenyl]propane (bis-GMA), 1,4-butanediol diacrylate (BUDA) or 1,6-hexanediol diacrylate (HDDA), EGDMA, and ECA. In Singapore, unpublished retrospective data showed that five allergens would have picked up all our past patients. These were EGDMA, 2-HPA, TREGDA, 2-hydroxyethyl acrylate (2-HEA), and urethane dimethacrylate. From such data, we found that only two allergens were in common in both centres. Hence, no single abbreviated series could be recommended for different centres.

In a recent article on dental personnel from Finland (8), 2-HEMA was found to be the most important allergen in dentists and dental nurses, and MMA and EGDMA in dental technicians.

In our present study, it is interesting to note that there was a considerable difference between the two centres in the intensity of positive reactions to the acrylate/methacrylate allergens studied. Of a total of 64 positive reactions (50 in Malmö and 14 in Singapore), the ++:++:+++ ratio in Malmö was 29:15:6, while the ratio in Singapore was 14:0:0. If we only consider the three main allergens and only patients who had undergone both readings, after subclassifying the patients as described in the *Materials and Methods* section, there was a significant difference between the two centres ($P = 0.005$, Mann–Whitney U -test).

In our previous studies in Malmö (3, 4), we had noted that all patients who reacted to EGDMA also reacted to 2-HEMA and that in every subject

with simultaneous positive patch tests to 2-HEMA and EGDMA, the strength of the positive reaction to 2-HEMA was either greater than or equal to the strength of the positive reaction to EGDMA. The same has been seen in the present study, where 2/9 (22%) of the simultaneous reactors showed stronger reactions to 2-HEMA and 7/9 (78%) the same reactivity. As noted in one of our previous articles (3), 2-HEMA can be present as an impurity in EGDMA, and EGDMA can be present as an impurity in 2-HEMA. Furthermore, EGDMA can be metabolized to 2-HEMA in the skin and body by enzymatic hydrolysis in cells and plasma, but 2-HEMA cannot be metabolized to EGDMA. This may explain the frequent simultaneous reactions to 2-HEMA and EGDMA.

However, in contrast to both our retrospective studies, the present study had one patient in Malmö who had reacted strongly to EGDMA only, without any simultaneous reaction to 2-HEMA. In contrast, among the Singapore patients in the present study, there were five patients who reacted to EGDMA, four of whom did not have simultaneous reactions to 2-HEMA. The absence of simultaneous positive reactions to 2-HEMA in these cases may be because the epitope for EGDMA in these cases was different from the postulated common epitope for both 2-HEMA and EGDMA in those cases with simultaneous reactions to both allergens, thus giving a different reaction product with endogenous proteins. An alternative explanation may be that these EGDMA-positive and 2-HEMA-negative reactions were due to acrylate or non-acrylate contaminants, which may have been presented only in the commercial EGDMA allergens. We must also consider the remote possibility that in this patient, the syringes for 2-HEMA and EGDMA might have been mixed up during the application of allergens to the Finn chambers®.

It is also interesting to note that although 2-HPA was the second commonest allergen in our retrospective data of Singapore patients, there was not a single positive reaction among all the patients tested to this allergen in the baseline series in the present prospective study. Furthermore, only three patients in Malmö had positive reactions to 2-HPA in the retrospective data, while 11 patients were positive to this allergen in the present prospective study, making it the third commonest acrylate/methacrylate allergen with positive results in this study.

Based on the present study with 38 positive reactors, if 2-HEMA alone had been used as a screening allergen, 21 (55%) positive reactors would have been missed. With 2-HEMA and TREGDA, only six (16%) would have been

missed, i.e. there would be a pickup rate of 84%. Those missed were four EGDMA-positive and two MMA-positive patients. The addition of EGDMA would give a pickup rate of 97%. All patients who had reacted to 2-HPA had simultaneous reactions to at least one of the other four allergens. In order to minimize the number of test preparations, a mix could be formulated. From our results in this study, we suggest that a mix of 2-HEMA, TREGDA, and EGDMA could be formulated into a screening acrylate/methacrylate mix. However, we must be aware that methacrylates and acrylates like 2-HEMA, TREGDA, or EGDMA cannot be used to screen for contact allergy to cyanoacrylates or epoxy acrylates.

In conclusion, whether acrylates/methacrylates should be added to the European and other baseline series will depend on whether the contact allergy rate in patients with suspected contact allergy to these allergens exceeds 0.5–1% (9) and whether a suitable mix of up to four acrylates can be formulated. Furthermore, care must be taken when formulating this mix, such that it must not actively sensitize patients at patch testing.

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