차량수리업에서 사용하는 이소시안계 페인트에 의한 피부와 눈의 노출에 관한 연구

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A Study of Dermal and Ocular Exposure to Isocyanate-Based Paints in Crash Repair Workshops

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Abstract : Exposure to HDI(hexamethylene di-isocyanate) commonly used in vehicle crash repair workshops remains a leading cause of occupational asthma. Although skin and eye contamination are considered as absorption routes, there are no occupational exposure standards for skin and ocular exposure. This is the reason why there are more empirical data should be provided. Therefore this study was to determine contamination levels of HDI on the skin, eyes, work surfaces, respirators and eye protectors. There was evidence of contamination on a variety of work surfaces, for example, door handles, bench top and spray gun, etc. A high proportion(47~80%) of skin wipe samples from neck, forehead, back hand, palm and wrist was positive for HDI contamination, even though spray time was relatively brief. The contamination levels from spraying inside spray booth were generally higher than outside booth due to poor work practices and inappropriate personal protective use like safety gloves. Apprentices had higher exposure levels than the qualified painters, likely due to lack of the recognition of safety and hygiene. The extent of contamination inside the PPE might provide an indication of the potential for respiratory & skin exposure and ocular exposure. Eye fluid samples from 4 out of 14 workers had the positive detection of HDI contamination, due to poor work practices like no or inappropriate eye protection. Considering the potential for dermal & ocular exposure to contribute to possible health symptoms including respiratory sensitization, the empirical data point to a need for improving work practices and appropriate PPE selection, use and maintenance.

초 록 : 차량수리작업장에 일반적으로 사용하고 있는 HDI의 노출은 직업성 천식을 발생시키는 주요 원인이 된다. 비록 피부나 눈의 오염이 흡수경로로 고려되고 있음에도 불구하고 이들의 폭로에 관한 작업장의 노출허용 기준이 규정되어 있지 않으므로 많은 실험 자료들이 제공되어져야 한다. 따라서 이 연구는 피부, 눈 그리고 호흡보호구와 눈보호구의 표면에 대한 HDI의 오염정도를 확인하였으며, 손잡이, 작업대 그리고 분사건과 같이 다양한 오염표면이 있다는 것도 명확히 하였다. 상대적으로 짧은 시간의 분사임에도 불구하고 목, 이마, 손등, 팔과 손목 등에서 채취한 시료의 많은 부분(47~80%)이 HDI오염에 양성이었다. 분사실내부에서의 오염정도는 좋지 못한 작업여건이나 보호경과 같은 부적절한 개인보호구 때문에 분사실외부의 오염정도보다 일반적으로 높다. 경험이 있는 페인트 공보다 초보자들의 폭로수준이 높은 것은 안전이나 보건의식의 결여 때문인 것으로 사료되고, 보호구내부의 오염정도는 호흡기관, 피부 및 눈의 잠재적인 노출을 야기할 것이다. 작업자 14명중에서 4명의 눈물시료로부터 HDI양성반응을 보였으며, 이것은 부적절한 눈 보호구에 의한 것으로 나타났다. 호흡기의 과민반응을 포함한 건강징후에 관여되는 피부와 눈의 노출 잠재성을 고려해볼 때 실질적 자료는 작업자들의 근무여건개선, 적절한 보호구의 선택과 사용 그리고 관리의 중요성을 잘 보여 주고 있다.
Key Words : crash repair workshops, dermal exposure, ocular exposure, Hexamethylene di-isocyanate, surface wipes

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1. Introduction

Organic isocyanates are present in the hardener component of the “two-pack” polyurethane spray paints commonly used in auto refinishing for crash repair. Aliphatic isocyanate oligomers, based on hexamethylene diisocyanate(HDI), are typically employed for so-called “clear coat” applications. Initially, volatile monomeric diisocyanates were used, but in the 1960s less volatile polyisocyanates were introduced to reduce inhalational exposure for non-spraying activities[1].

However, despite the efforts of reducing isocyanate exposure several decades of research and intervention, exposure to isocyanates in crash repair workshops remains a leading cause of occupational asthma[2]. Based on animal evidence, it has been suggested that skin contact with isocyanates may lead to asthma[3,4], emphasizing the importance of the control of dermal exposure.

It has also been suggested that contaminated working surfaces and poorly controlled skin exposure to isocyanates might be associated with respiratory symptoms[5-10]. Biological uptake in humans may also be caused by skin absorption or other exposure routes[11]. Previous studies in crash repair workshops have identified an increased prevalence of skin problems, such as dry cracked skin, dermatitis, skin irritation[5,9,10]. From these studies, the possible adverse health symptoms were related to skin or surface contamination resulting from inappropriate personal practices and personal protective equipment. However, it was commonly recommended that there should be more empirical data related to skin & surface contamination. In particular, there are no occupational exposure standards for skin exposure. Although ocular exposure can be an exposure route like dermal and inhalational exposure, there is no available data for ocular contamination, because of a lack of attention of ocular exposure, the significance of this route being unclear and no standardized occupational exposure monitoring methods available. Therefore, this study was conducted to determine skin & work surface contamination including protective equipment and possible ocular exposure to isocyanate.

2. Materials and Methods

2.1. Study Population

Twenty-six spray painters from nine commercial automobile repair shops including four apprentices from a TAFE(Technical and Further Education) college and a Motor Trade Association(MTA) training school, and three outdoor spray painters participated in this study. They were largely drawn from individual small businesses that were members of MTA.

Workers appeared to have repeated exposure via aerosol deposition on exposed skin or clothing. Respirator, clothing and shoe contamination were often visible, despite the common use of high volume low pressure spray guns. The PPE was washed infrequently and often stored inappropriately, resulting in persistent surface contamination as reported by a previous study[9].

Workers were also observed to touch contaminated surfaces of PPE and spray guns with their hands without wearing protective gloves. The mixing rooms, bench tops and floors were potential sources of contamination, due to chemical spillage. Small spray painting tasks sometimes occurred outside of the dedicated booths increasing the likelihood of skin/surface contamination.

2.2. Monitoring Methods

Surface Monitoring : IPA(isopropanol) was sprayed on the contaminated surface like chemical balance, bench top, rocker handle, door handle and spray gun and paper tape(aliphatic isocyanate) and/or *Swype* indicators were used on a small area for screening purposes[12]. Wiping area was 10cm×10cm or whole area of door handles, rocker handle and a spray gun handle. After the detection of isocyanate contamination, for quantitative assessment the surface was re-sprayed and wiped with a Ghost wipe pad, and treated as below.

Dermal Monitoring : For qualitative assessment of skin contamination, commercial products, namely *Swypes* and *Permea-Tec* pads(Omega Speciality Instrument Company, USA) were used[11]. A color changed from yellow to orange appeared to indicate the presence of aliphatic isocyanates. For quantitative dermal monitor-

ing, a small amount of pure IPA was sprayed on a whole area of body regions like neck, forehead, hands, palms and wrists, and then the sprayed regions were wiped by *Ghost* wipe pads[9] purchased from Environmental Express(USA). The wipes were treated with the derivatising solution(see below) prior to analysis.

PPE Monitoring : Surface wiping of PPE was carried out using *Ghost* wipe pads and IPA. For this investigation the inside and/or outside surfaces of PPE were wiped immediately after paint spraying activity.

Ocular Monitoring : Commercial eye drop solution ("Refresh", *Allergan*) was used for the ocular monitoring. In laboratory experiments to determine the stability of HDI in the commercial eye drop solution, 10μL of hardener solution(5.7% of NCO group in liquid hardener, PPG, 2K MS Normal hardener) was dissolved in 1.5mL of eye drops. The HDI-based hardeners were found to break down relatively quickly in the eye drop solution, with only about 10% of the isocyanate remaining after 2min. at room temperature.

Therefore, ocular sampling for HDI was carried out in workers immediately(less than 2 min.) after spraying. Ocular fluid samples were collected by applying 0.1~0.2mL of the commercial eye drops into each eye. Excess liquid from the corner of each eye was absorbed onto a sterile cotton swab, and this was treated as below in analytical methods.

2.3. Analytical Methods

During the sampling, clean disposable nitrile gloves were worn and tweezers were used to pick up the pads to minimize contamination by hands or surfaces. Isocyanate-containing samples in wipes and swabs were placed in vials containing 10mL of the derivatising solution dissolved 50μg of 1-2MP(methoxy-phenyl piperazine) in 1mL of dried toluene[13]. Sampling vials were stored in an ice box prior to transport to the laboratory. After 24 hours, 200μL of acetic anhydride was added into the vials, and left for 30min. Evaporation of the samples was carried out in a fume cupboard by blowing pure nitrogen gas. In order to re-dissolve the residue, 10mL of acetonitrile was added. In the case of the eye swab samples, 5mL was used. The adjustment of the results from the samples was carried out using a blank sample.

Analysis was by HPLC(*Spherisorb* ODS2(C18) column, +0.8V EC, 242nm UV, 30°C oven temperature) in accordance with a previous study[7]. The mobile phase comprised 67% acetonitrile, 33% distilled water and pH 6.0 controlled by sodium acetate and acetic acid. Flow rate was 1.5mL/min with helium sparging.

The limit of detection was 0.003μgNCO/mL for the EC detector(0.008 μgNCO/mL for the UV detector). The sensitivities of the paper tape and *Swype* indicators were approximately 0.002μgNCO/mL. Retention times of monomeric and polymeric HDI are 3.1 and 7.8minutes respectively.

3. Result and Discussion

3.1. Surface Monitoring

Table 1 provides qualitative data on surface contamination, and table 2 gives the quantitative results. The limit of isocyanate detection is 0.03μgNCO in total. The abbreviations in table 1, 2 and assessment areas are described as CB: chemical balance(100 cm^2), BT: bench top(100cm^2), RHM: rocker handle in mixing room(approx. 66cm^2), IDHM: inside door handle in mixing room(typically 70cm^2), ODHM: outside door handle in mixing room(typically 70cm^2), IDHB: inside door handle in spray booth(approx. 98cm^2), ODHB: outside door handle in spray booth(approx. 98cm^2) and SG: spray gun(approx.100cm^2).

The interpretation of colour change data in table 1 is similar to that of a previous study[8]. That is, a color

Table 1. Isocyanate indicator testing of surfaces using paper tape or Swype pads

Workplace i.d.	Colour reaction (P = positive, N =negative)					
	CB	BT	IDHM	ODHM	IDHB	ODHB
B1	P	P	P	P	P	P
B2	P	P	P	P	N/S	N/S
B3	P	N/S	P	P	P	P
B4	N[a]	P	P	P	N/S	N/S
B5	P	P	P	N[b]	N/S	N/S
B6	P	P	P	P	N/S	N/S
B7	P	P	P	N[b]	N/S	N/S
B8	P	P	N[b]	N[b]	N/S	N/S
B9	P	P	P	P	P	P

a: Cleaned after use, b: Open all the time without touching, N/S: Not samples

Table 2. Quantity of isocyanate on surface samples in spray and mixing areas

Workplace i.d.	Total isocyanate (μg NCO)							
	CB	BT	RHM	IDHM	ODHM	IDHB	ODHB	SG
B1	0.05	< 0.03	< 0.03	< 0.03	< 0.03	42	< 0.03	21
B2	< 0.03	N/S	N/S	< 0.03	< 0.03	N/S	N/S	N/S
B3	N/S	N/S	N/S	0.1	1.2	N/S	N/S	0.4
B4	< 0.03	< 0.03	N/S	< 0.03	< 0.03	1.6	0.5	11
B5	< 0.03	N/S	N/S	0.04	< 0.03	N/S	N/S	0.2
B6	0.12	N/S	N/S	0.04	0.04	N/S	N/S	0.3
B7	< 0.03	< 0.03	N/S	< 0.03	< 0.03	0.3	0.1	< 0.03
B8	< 0.03	< 0.03	N/S	< 0.03	< 0.03	< 0.03	< 0.03	< 0.03
B9	N/S	0.05	N/S	0.9	N/S	N/S	N/S	4.6

* N/S: Not samples

changed from yellow to orange appeared to indicate the presence of aliphatic isocyanates(i.e. -NCO group).

Table 2 illustrates the large variability in surface contamination, potentially due to poor work practice, initial isocyanate strength, partial polymerization and other factors[8]. Most values(57%) were below the limit of detection(0.03μgNCO) with 30% between 0.04 and 1.0μgNCO, and 13% exceeded 1.0μgNCO. The highest recorded values were 21 and 42μgNCO, both swabs collected from business B1 mainly due to infrequent cleaning process and frequent tasks.

The amount and distribution of surface contamination is likely to be highly variable, due to different mechanisms like direct contact, aerosol deposition and surface contaminant transfer, isocyanate strength, extent of isocyanate reaction and other factors[14,15]. Furthermore, the amount of surface contaminant removed by wiping is unlikely to be uniform across all surfaces[8,16], and therefore the results presented here should be considered semi-quantitative.

3.2. Dermal Monitoring

Skin wipe results for the painters spraying inside and outside booths are given in table 3. The limit of isocyanate detection is 0.03μg NCO in total. The abbreviations in table 3 are described as N: neck, FH: forehead, LBH: left back hand, RBH: right back hand, LP: left palm, RP: right palm, LW: left wrist and RW: right wrist, also the alphabetical symbols are expressed as a: no protection, b: wore disposable nitrile gloves, c: covered by disposable overall, d: wore disposable latex gloves, e: touched by contaminated hands, f: wore disposable coverall and g: wore air line hood.

It can be seen that the apprentices (S1,7,11,12) have somewhat higher exposure levels than the qua-

Table 3. Skin wipe data for spray painters

Person i.d.	Activity duration (minutes)	Total isocyanate (μg NCO)							
		N	FH	LBH	RBH	LP	RP	LW	RW
Inside booth									
S2	1	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{c}$	< 0.03 c
S3	2	$< 0.03^{a}$	$< 0.03^{a}$	0.06^{d}	$< 0.03^{d}$	0.08^{d}	$< 0.03^{d}$	$< 0.03^{a}$	$< 0.03^{a}$
S4	3	0.08^{a}	0.09^{a}	0.17^{a}	$< 0.03^{a}$	0.08^{a}	0.11^{a}	2.0^{a}	$< 0.03^{a}$
S5	4	$< 0.03^{a}$	$< 0.03^{a}$	0.05^{a}	0.04^{a}	2.0^{a}	0.11^{a}	0.14^{a}	0.06^{a}
S6	4	0.09^{e}	0.11^{e}	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{a}$
S8	7	$< 0.03^{f}$	$< 0.03^{f}$	$< 0.03^{d}$	$< 0.03^{d}$	0.05^{d}	0.04^{d}	0.04^{a}	$< 0.03^{a}$
S9	9	0.31 e	$< 0.03^{e}$	$< 0.03^{d}$	0.19^{d}	0.18^{d}	$< 0.03^{d}$	0.06^{a}	0.08^{a}
S10	15	0.13^{a}	0.30^{a}	0.08^{a}	0.05^{a}	0.07^{a}	0.15^{a}	0.12^{a}	0.27^{a}
S1*	1	$< 0.03^{f}$	$< 0.03^{f}$	0.25^{a}	0.18^{a}	1.2^{a}	0.72^{a}	$< 0.03^{f}$	$< 0.03^{f}$
S7*	5.3	0.79^{f}	0.11^{f}	0.15^{d}	0.1^{d}	0.42^{d}	0.39^{d}	0.5^{a}	0.17^{a}
S11*	25	0.2^{a}	$< 0.03^{g}$	$< 0.03^{d}$	0.48^{d}	0.1^{d}	0.48^{d}	0.43^{a}	3.1^{a}
S12*	30	1.5^{a}	2.5^{a}	2.5^{a}	2.6^{a}	1.7^{a}	2.2^{a}	2.7^{a}	2.1^{a}
Outside booth									
S13	2.4	0.54^{a}	0.19^{a}	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{b}$	$< 0.03^{a}$	0.06^{a}
S14	7.3	0.35^{a}	0.54^{a}	0.80^{a}	1.08^{a}	1.83^{a}	2.58^{a}	$< 0.03^{a}$	$< 0.03^{a}$
S15	31	$< 0.03^{a}$	$< 0.03^{a}$	0.08^{a}	0.07^{a}	0.06^{a}	$< 0.03^{a}$	$< 0.03^{a}$	$< 0.03^{a}$

lified painters. Clothing and gloves afforded protection, except for those using disposable latex gloves. Screening with skin *Swypes*, underneath the disposable latex, yielded a color change.

It appeared that spraying outside of the booth, i.e. in open areas, as expected, does not have higher dermal exposure levels than spraying inside the booth. However, S14 had appreciable skin exposure, due to proximity to the object being sprayed and both S13 and S14 had measurable contamination on the neck and forehead. A high proportion of skin wipe samples tested positive, even though spray time was relatively brief. Sampling protocols for dermal exposure[17] should include body locations close to the spray, such as neck and forehead areas.

Biological monitoring like hexamethylene diamine (HAD) as a biomarker of HDI based compounds of isocyanate exposure will assist in understanding the extent of uptake once skin, eye or inhalation exposure occurs[18,19].

Table 4 summarizes the proportion of positive skin results by body regions from 15 spray painters out of 26 in total. It can be seen that the majority of skin wipes yielded a positive result, even though spray time was relatively short.

3.3. PPE Monitoring

Results for wipes of PPE are given in table 5 and 6. The limit of isocyanate detection of those is also 0.03 g NCO as before, and S1, S7 and S12 were apprentices.

The abbreviations in table 5, 6 and assessment areas are described as SIAR: inside transparent surface of hood air line respirator(approx. 560cm^2) SOAR: outside transparent surface of hood air line respirator (approx. 560cm^2), SIR: inside surface of air purifying half face respirator(approx. 60cm^2), IG: inside safety goggle(approx. 56cm^2) and OG: outside safety goggle(56cm^2). Also the alphabetical symbols are expressed as a: not cleaned before and after use, and stored in contaminated area: b: poor facial fit, due to beard and different size, c: touched by contaminated hands, and stored in contaminated area.

Table 4. Proportion of detectable dermal isocyanate exposures by body location

Body location# (n=15)	% positive
Neck	60
Forehead	47
Left back hand	60
Right back hand	60
Left palm	80
Right palm	67
Left wrist	53
Right wrist	47

Table 5. Isocyanate contamination of PPE for painters spraying in a booth

Person i.d.	Total isocyanate (μg NCO)		
	SIAR	SOAR	SIR
S2	2.8[a]	21.8	-
S3	N/S	N/S	2.60[a]
S4	N/S	N/S	0.08[b]
S5	N/S	N/S	< 0.03[b]
S6	N/S	N/S	< 0.03[b]
S8	N/S	N/S	0.07[a]
S9	N/S	N/S	0.86[a]
S10	< 0.03[b]	< 0.03	N/S
S1*	0.44[a]	9.0	N/S
S7*	1.61[c]	9.0	N/S
S12*	< 0.03	< 0.03	N/S

* N/S: Not sampled

Table 6. Isocyanate contamination of PPE for painters spraying outside of a booth

Person i.d.	Total isocyanate (μg NCO)		
	IG	OG	SIR
S13	0.14[a]	0.97	0.53[a]
S14	-	-	1.17[a]
S15	0.07[a]	< 0.03	< 0.03[a]

It appears that inside surface contamination, when detectable, is approximately an order of magnitude less than external surface contamination.

3.4. Ocular Monitoring

A limited amount of eye contamination sampling was conducted. The results are given in Table 7. It appears that eye protection tools like hood or safety goggles are effective. However, touching eyes with contaminated hands after the spray painting or not wearing eye protection resulted in eye contamination.

Table 7. Ocular isocyanate exposure for spray painters

Location		Eye protection	Range of exposure (μgNCO)	
			Left eye	Right eye
Inside booth (n=11)	Qualified painters (n=7)	Yes(air line hood) (n=3)	n.d., n.d., n.d.	n.d., n.d., n.d.
		NO (n=4)	n.d., n.d., n.d., 0.02	n.d., n.d., n.d., 0.03
	Apprentices (n=4)	Yes(air line hood) (n=3)	n.d., n.d., 0.1*	n.d., n.d., 0.18*
		NO (n=1)	0.05	0.25
Outside booth (qualified painters) (n=3)		Yes(safety goggles) (n=2)	n.d., n.d.	n.d., n.d.
		NO (n=1)	0.05	0.02

* Observed to touch eyes with contaminated hands after spraying.
n.d. = not detected < 0.02μg NCO in total

This is the first study reporting a measurement of isocyanates in eye fluid. Eye fluid samples were positive for -NCO in samples from 4 workers of 14 tested. When detected, -NCO was present in samples from both eyes in each case. Given the reactivity of isocyanates, it was a somewhat unexpected finding, but some degree of ocular contamination is likely to occur by not wearing any, or inappropriate, eye protection or indirect transfer via contaminated hands. The apprentices appeared to have been exposed to more isocyanate than the qualified painters, perhaps due to a lesser awareness of exposure routes.

Even though some ocular exposure was evident, no excess of eye symptoms was reported, consistent with a previous study[9]. However, ocular sampling methodologies should be further developed and there is a need to better understand the significance of the ocular route, and the influence of wearing contact lenses.

4. Conclusion

Dermal and ocular exposure to isocyanate in crash repair workshops were investigated in this study. Especially, this is the first study reporting a measurement of isocyanates in eye fluid. The results we have obtained are as bellows :

1) The amount and distribution of surface contamination are likely to be highly variable, due to different mechanisms. However, the use of appropriate PPE reduced skin and eye contamination, which may reduce the possibility of dermatitis and respiratory symptoms although there is a lack of evidence that dermal contact causes respiratory sensitization.

2) Even though some ocular exposure was evident, no excess of eye symptoms was reported. Eye fluid samples were positive for -NCO in samples from 4 workers of 14 tested. When detected, -NCO was present in samples from both eyes in each case. However, ocular sampling methodologies should be further developed and there is a need to better understand the significance of the ocular route, and the influence of wearing contact lenses.

3) There is a need to educate workers about minimizing exposure via the dermal and ocular routes via appropriate PPE and work practices like cleaning. Because the apprentices appeared to have been exposed to more isocyanate than the qualified painters, perhaps due to a lesser awareness of exposure routes.

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