Evaluation of the Efficiency of Respiratory Protective Equipment based on the Biological Monitoring of Styrene in Fibreglass Reinforced Plastics Industries

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Abstract: Evaluation of the Efficiency of Respiratory Protective Equipment based on the Biological Monitoring of Styrene in Fibreglass Reinforced Plastics Industries: Shoji Nakayama, et al. Department of Public Health, Okayama University Graduate School of Medicine and Dentistry—The purpose of the present study was to determine the efficiency of respiratory protective equipment in a fibreglass reinforced plastic factory by comparing results of environmental and biological monitoring of exposure to styrene. Five factories including 39 workers were investigated. Three types of respiratory protective equipment were tested: one was a half-mask air-purifying respirator equipped with a cartridge for organic solvents, another was a disposable gauze respirator impregnated with charcoal filter, and the third was a dust-proof respirator. The frequency of cartridge exchange of a half-mask respirator was twice a day only at one factory, and that was less than once a month at other factories. The site concentrations exceeded 20 ppm at 10 of the 82 sampling points (12.2%), and 22 of the 39 workers’ personal exposure exceeded 20 ppm which is the current occupational exposure limit recommended by the Japan Society for Occupational Health. The efficiency of disposable gauze respirators and dust-proof respirators was low or rather zero. The average efficiency of half-mask respirators in which cartridges were exchanged twice a day and once a month was 83.6% and 46.6%, respectively. There was a significant disparity in the efficiency of the respirator depending on the frequency of cartridge exchange (p < 0.05). Overall this study showed that even though a half-mask respirator is used and its cartridge is exchanged every half a day, workers exposed to a styrene concentration at or over 122 ppm are expected to inhale more than 20 ppm of styrene. (J Occup Health 2004; 46: 132–140)

Key words: Efficiency, Protective equipment, Biological monitoring, Styrene, FRP, Cartridge, Exchange frequency

Over the past few decades, a considerable number of studies have been conducted on occupational exposure to styrene and its various health effects. Health problems include central and peripheral nervous system disorders, dermatitis, mucous membrane irritation, liver and kidney disorders, respiratory symptoms, and haematological abnormalities¹⁻⁵. It has also been reported that reproductive, genetic, and mutagenic toxicity were observed on styrene exposure⁶⁻¹⁰. In addition, there has been growing concern over the last several years about the impairment of colour vision and healing ability even at a relatively low dose of exposure¹¹⁻¹⁵. Based on these reports, the Japan Society for Occupational Health recommended 20 ppm or less as the occupational exposure limit (OEL) for styrene in 1999¹⁶.

Styrene monomer is one of the most important materials used to produce plastics and rubbers. It is reported that the highest exposure to styrene is found in the manufacture of fibreglass reinforced plastics (FRP)¹⁷⁻¹⁸. FRP products include fishing boats, water tanks, septic tanks, shower stall and bathtub units, built-in kitchen tables, ducts for power supply lines, and so forth.

Styrene vapours should preferably be reduced by engineering efforts, such as changes in production processes or the introduction of appropriate ventilation...
systems. But in most cases, factories manufacturing FRP are on a relatively small scale in Japan and have a difficulty introducing such measures. Using personal protective equipment such as half-mask respirators might often be the only way of reducing the intake of styrene in these workshops.

The purpose of the present study was to estimate the efficiency of respiratory protective equipment based on the biological monitoring in FRP industries. The estimation was done by comparing workers’ personal exposures to styrene with its metabolite in urine samples collected at the end of the work shift. We also evaluated the influence of the frequency of cartridge exchange in half-mask respirators on their efficiency, since there are few reports that discuss this issue.

Materials and Methods

Subjects

Thirty-nine workers from five factories in Okayama, Japan, participated in this study. None of the subjects had an abnormality in the specific health examination for organic solvents. These factories produce fibreglass reinforced polyester plastics with a material containing styrene monomer in various proportions.

The subjects ranged in age from 21 to 68 yr (44.5 ± 11.3 on average ± standard deviation). Factory A with 14 subjects (42.2 ± 15.7 yr old), had two plants, one that manufactured FRP products such as water/septic tanks and shower stall and bathtub units, and another that produced reinforced plastics such as fitted (built-in) kitchen counter tables and tank shells using press machines. Factory B, with 6 subjects (54.3 ± 11.9 yr old), manufactured water-purification tanks, septic tanks, oil trap tanks, silos, and so on. Factory D, 11 subjects (44.9 ± 6.2 yr old), manufactured FRP boats in a very large workplace. Factories C and E, 3 and 5 subjects (43.2 ± 13.1 and 42.8 ± 13.6 yr old, respectively, handled block or sheet material compound containing a styrene and fibreglass mixture to press reinforced polyester plastic products.

All subjects gave their consent after being fully informed about this study, its purpose, its benefits for occupational health, and their right to withdraw from the study at any time.

Inspection

At the beginning of this study, we inspected the workplaces to investigate the working processes, the working environment, and the type of protective equipment and how it was worn. All subjects using half-mask respirators were asked about the frequency of respirator cartridge exchange.

Site concentration and personal exposure dose

The styrene concentration in workers’ ambient air (site concentration) was measured in accordance with the Japan Standard for Working Environment Measurement. We divided workplaces into 3–5 m mesh and each sampling point was located on the mesh. Five litres of air were sampled in Tedlar® Bags (GL Sciences Inc.) on Friday in each factory. Styrene concentrations were determined by gas chromatography and a flame ionisation detector (GC-FID, Hitachi) with a PEG 600 column (GL Sciences Inc.). Instantaneous concentrations of styrene during the various working processes in several workplaces in factory D were measured with a portable gas detector (XP-316, NEW COSMOS ELECTRIC Co. Ltd, Japan, Fig. 2).

The personal styrene exposures were measured by personal air sampling in the workers’ breathing zone. A passive sampler (Organic Vapour Monitor 3500, 3M) was attached to the collar of each worker at the beginning of the work shift. The worker was asked not to take it off until we removed it. Each sampler was collected and sealed in an aluminium container, and the precise time of the sampling was recorded. All samplers were carried back to the laboratory, extracted with carbon disulphide (Wako Pure Chemical), and the styrene concentration was determined by GC-FID (Hitachi) with a DB-WAX column (GL Sciences Inc.). The eight-hour time weighted average concentration (8 h-TWA) was calculated to indicate personal exposures to styrene.

Measurement of urinary mandelic acid

Urine samples were collected from all subjects at the end of the work shift, put in an ice-cooled container, carried back to the laboratory, and stored in a refrigerator until analysis. The concentration of mandelic acid (MA), the major metabolite of styrene, and creatinine were determined simultaneously by high performance liquid chromatography (HPLC, Hewlett-Packard) with Hypersil ODS (Hewlett-Packard) as a column\textsuperscript{19}. The concentration of urinary MA was corrected with one gram of creatinine per litre for the analysis.

Estimation of the efficiency of the respirators

We estimate the concentration of styrene that passed through the respirator, namely inhaled styrene, from the excretion of mandelic acid in urine. This value was compared with the personal exposure dose, and the efficiency of the respirator was calculated.

In this study, we adopted the previous ACGIH recommendation, 800 mg/g · creatinine for the BEI (biological exposure index) of the urinary MA concentration corresponding to 50 ppm for TLV-TWA (threshold limit value as the time weighted average concentration) of styrene exposure. Assuming the relation between inhaled styrene and excreted MA, the estimated inhaled concentration of styrene (ISTYR) was calculated according to the following equation:
ISTYR (ppm) = \frac{50 \text{ (ppm)}}{800 \text{ (mg/g · creatinine)}} \times \text{MA (mg/g · creatinine)}

where MA is the actual urinary MA concentration (mg/g · creatinine) at the end of the work shift. Then the fraction (F) of inhaled styrene to the personal exposure level (PE) was calculated as follows:

F (%) = \frac{\text{ISTYR}}{\text{PE}} \times 100

where PE is the personal exposure concentration (ppm). And the efficiency of respirators (E) must be

E (%) = 100 – F

Consequently, E was calculated by means of the following equation:

E (%) = 100 – 6.25 \times \frac{\text{MA}}{\text{PE}}

Statistical analyses

Group mean comparisons were performed with the Kruskal-Wallis H statistic followed by Scheffé’s method. A significance level of 0.05 was used for all statistical tests, and two-tailed tests were applied.

Results

Inspection of a workplace

The type of respirator was different for each plant, and the following three types of respirators were used. One was a half-mask air-purifying respirator equipped with a cartridge for organic solvents (half-mask respirator), another was a disposable gauze respirator impregnated with charcoal filter (gauze respirator), and the third was a dust-proof respirator (dust respirator). These respirators are shown in Fig. 1. All workers wore rubber protective gloves.

Some workers answered that they sometimes did not wear respirators and used only dust respirators for the grinding and finishing procedure. Respirators and their cartridges were supplied by the employer and the type of respirator that should be used in the workplace was determined by the employer, but whether and how to wear the respirator was left to the workers’ discretion; the most common reason why workers did not wear respirators was that they felt discomfort and had difficulty breathing. Incorrect respirator use was observed in some plants, such as slipping a face-piece out of the correct position or putting gauze between the face and the face-piece to make breathing easier. The frequency of cartridge exchange for a half-mask respirator was different for each factory: twice a day in factory D, and less than once a month in the other factories. As a matter of fact, the workers as well as employers did not know when they should replace the cartridge. No local air exhaust system, except for a general ventilation system, was introduced in each plant manufacturing such large products as fishing boats or relatively small products such as water tanks. Although the spraying process was performed in a ventilation booth in factory A, some workers with gauze respirators moved to the leeward side of the products when spraying.

Site measurements, personal measurements, and calculation of respirator efficiency

Table 1 shows results of measurements and calculations, jobs, the type of respirator, and the frequency of cartridge exchange in half-mask respirators. Styrene concentrations at the mesh sampling points closest to workers were assigned to site concentrations for the workers.

Instantaneous concentrations for several work processes in factory D ranged from 10 to 2,000 ppm, and the values varied widely from process to process. A gelcoating process included the spraying of a gelcoating agent containing 40%–50% styrene, presented an extremely high concentration of 2,000 ppm. A fibreglass/resin (30%–40% styrene) spraying process also showed high concentrations (Fig. 2). Concentrations in the

Fig. 1. Three types of respiratory protective equipment. A: half-mask air-purifying respirator, B: disposable gauze respirator impregnated with charcoal filter, C: dust-proof respirator.
Table 1. Site concentration during the workshift, personal exposure (8-h TWA), urinary MA at the end of the workshift, and respirator efficiency

<table>
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<th>Worker</th>
<th>Site concentration (ppm)</th>
<th>Personal exposure (PE, ppm)</th>
<th>Mandelic acid in urine (MA, mg/g-creatinine)</th>
<th>Respirator efficiency (E, %)</th>
<th>Job</th>
<th>Type of respirator</th>
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*: Calculated as E = 100 – 6.25MA/PE, †: ND<0.5 (ppm)
breathing zone of laminators were 100–200 ppm. On going away from the product, the styrene concentration decreased rapidly. The concentration was about 300–500 ppm on the products, but was 100–200 ppm at a place 2–3 m away, and below 20 ppm, TLV-TWA, farther away.

Discussion

Both site concentrations and personal exposures were highest in factory D. This may be ascribed to differences in working processes. About 10 m long fishing boats were manufactured in factory D. Gelcoat agent and fibreglass/resin mixture were scattered around the workplace (Fig. 2), so that the styrene concentration in workplace air reached very high concentrations.

There were 6 of the 26 sampling points (23.1%) where the site concentrations exceeded 20 ppm in factory D. Three of the 11 points (27.3%) and 1 of the 11 points (9.1%) were also above 20 ppm in factory B and factory E, respectively. Altogether, 10 of the 82 sampling points (12.2%) exceeded the OEL. On the other hand, 22 of 39 workers (56.4%) had the personal exposures above 20 ppm (Table 1). There was at least one worker whose exposure exceeded the OEL in each factory. These results indicate that the working environment in these factories should be improved to meet the OEL. They will not be able to meet the administrative level of styrene when it is reduced in future.

The FRP production process included gelcoating (spraying a surface paint and an agent to easily separate a product from a mould), spraying a fibreglass/resin mixture, lamination of a fibreglass sheet, impregnating and rolling to remove any air bubbles, hardening, attaching ribs, removing the mould, and grinding and finishing. The longest time in the whole FRP procedure was spent by workers in the hand laminating process so that personal exposures were mostly derived from site concentrations in this process. The gelcoating and spraying processes took a relatively short time, 10–15 min, during the entire work shift, although the processes included one of the highest exposure levels. In fact, no significant difference in personal exposure was found in a mean comparison between the laminator group and the sprayer group. The spraying process, more than 2,000 ppm, seems to have less effect on the average exposure, but an acute poisoning cannot be ignored. Measuring an instantaneous concentration with a portable gas detector is considered useful for monitoring the acute intoxication.

The plant that manufactured large-sized products such as a fishing boat was as large as a gymnasium and introduced a general ventilation system; the site concentrations therefore varied with the sampling points in the plant. The other plants producing smaller-sized products such as water tanks had a relatively smaller building and produced many products simultaneously in the building. Therefore the site concentrations were ubiquitously high. The styrene concentrations and exposures were higher on the products made in concave moulds such as boats and tanks, than on ones made in convex moulds such as bathtubs, because the lamination and handrolling procedures of the former were performed ‘inside’ the products while those of the latter were done ‘outside’. Workers in factory B were occasionally engaged in laminating inside a silo. Ikeda et al.20) reported that the installation of a local exhaust system with several flexible hoses is a very effective means in the boat
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manufacturing workshop. As mentioned above, styrene exposure should be reduced by these engineering modifications, but the factories are on a relatively small scale and have difficulty introducing such measures. Therefore using personal protective equipment will be one of the best ways of reducing the intake of styrene in these workshops.

The correlation coefficient between site concentrations and personal exposures was 0.652 ($p<0.05$). The equation for the regression curve was $y=10.59 + 3.38x$, where $x$ and $y$ represent the site concentration and the personal exposure, respectively. It was indicated that the personal exposure was higher than the site concentration from the slope of the regression line. Personal exposures were indeed significantly higher than site concentrations ($p=0.00001$) when the comparison was performed with the Wilcoxon’s matched pairs signed ranks test (two sided). The workers stooped down and put their upper body to the product during lamination.

Personal exposure was measured in the workers’ breathing zone, on the other hand, the site concentration was determined at a point away from the product. This is the reason for the difference between personal exposures and site concentrations. It is quite likely that the personal exposure reflects practical exposure better than the site concentration so we adopted the personal exposure in further analyses.

Differences between mean efficiencies in gauze and dust respirators were not significant ($p=0.988$). Thus we classified workers into three groups, i.e., the twice-a-day group in which workers wore half-mask respirators with cartridges exchanged twice a day, the once-a-month group in which workers wore half-mask respirators with cartridges exchanged less than once a month, and the third was the gauze-or-dust group in which workers wore gauze or dust respirators.

| Fabrik | Half-mask respirator | | | | | Halve-mask respirator | | | | | | Gauze or dust respirators |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Twice a day cartridge exchange | | Less than once a month cartridge exchange | | | | | | | | | | | |
| | Personal exposure (ppm) | Mandelic acid in urine (mg/g-creatinine) | Respiator efficiency (%) | Personal exposure (ppm) | Mandelic acid in urine (mg/g-creatinine) | Respiator efficiency (%) | Personal exposure (ppm) | Mandelic acid in urine (mg/g-creatinine) | Respiator efficiency (%) | |
| 18.0 | 70 | 75.7 | 10.0 | 24.1 | 84.9 | 0.7 | 65.1 | 0 |
| 66.3 | 110 | 89.6 | 10.2 | 432.9 | 0 | 1.6 | 81.8 | 0 |
| 83.8 | 390 | 70.9 | 14.6 | 222.5 | 4.8 | 3.4 | 95.5 | 0 |
| 84.8 | 270 | 80.1 | 15.3 | 151.6 | 38.1 | 8.0 | 110 | 14.1 |
| 203.5 | 310 | 90.5 | 17.8 | 88.0 | 69.1 | 14.3 | 171.7 | 25.0 |
| 203.9 | 680 | 79.2 | 24.6 | 192.4 | 51.1 | 16.8 | 184.9 | 31.2 |
| 212.3 | 790 | 97.9 | 26.7 | 272.5 | 36.2 | 17.8 | 136.5 | 52.1 |
| 283.7 | 420 | 90.7 | 28.3 | 809.0 | 0 | 17.9 | 399.8 | 0 |
| 318.8 | 960 | 81.2 | 29.2 | 130 | 72.2 | 24.9 | 204.4 | 48.7 |
| 31.1 | 153.4 | 69.2 | 31.1 | 153.4 | 69.2 | 39.7 | 361.2 | 43.1 |
| 32.7 | 60 | 88.5 | 40.5 | 549.9 | 15.1 |
| 37.4 | 662.9 | 0 | 43.5 | 483.4 | 30.5 |
| 54.3 | 390.7 | 55.0 | 72.2 | 1606.0 | 0 |

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Significant $a$ $p<0.05$ $b$

c $d$

d

Group mean comparisons were performed with the Kruskal-Wallis test followed by Scheffé’s method.
respectively. Since the correlation for two half-mask or dust respirator groups were 0.676, 0.326, and 0.898 (coefficients for the respirators was very low, or rather zero. Correlation results indicate that the efficiency of the gauze and dust respirator were excluded from the graph. The ACGIH relation, line D (y = (800/50)x + 116, R=0.898), was close to the ACGIH relation. Weber and Mullins found 39.7 for the geometric mean penetration ratio of the FRP workers. According to this equation, the penetration ratio was converted to 98.7% of the respirator efficiency. Weber and Mullins mentioned, there are some technical problems in determining the WPF. It is difficult to collect vapour inside a respirator cavity, to analyse the extremely low doses of the contaminants, and to deal with the bias of pulmonary retention and skin absorption. Therefore we tried to determine the adsorbed styrene dose by a relatively easy handled method, that is, biological monitoring. The urinary MA is measured routinely in Japan according to the Japan Ordinance on the Prevention
of Organic Solvent Poisoning. We adopted the urinary MA concentration as the index of styrene exposure. Of course, there are also some problems in this method. They include skin absorption, the variability of the excretion of MA, metabolic saturation, and accumulation of styrene to which exposed the previous day.

There are at least two studies that reported percutaneous absorption of styrene is not a significant exposure source and does not significantly affect the body burden of styrene in the FRP industry, so that we did not take the influence of skin absorption into account in this study.

The variability of styrene metabolism between and also within individuals is difficult to deal with. The correlation between personal exposures and the urinary MA in workers using gauze or dust respirators was relatively high, $R=0.898$, so that the variability in the excretion of MA was considered to be acceptable in this study.

Löf and Johanson pointed out that metabolic saturation occurred when the workers were exposed to concentrations of 100–200 ppm. Ikeda et al. also maintained that a linear relationship was found below a 150 ppm styrene concentration. Therefore it must be necessary to mention the linearity in the high styrene concentrations observed in our study. As shown in Table 2, workers in the twice-a-day group, i.e. factory D, were exposed to over 150 ppm of styrene. Therefore the metabolism of styrene was saturated so that the excretion of the urinary MA might be underestimated and the respirator efficiency might be overestimated. The problem can be solved by comparing the urinary MA when workers wear the respirator appropriately with the MA when they do not wear any respirator. If the ethical problem can be resolved we will be able to ask the workers not to wear the respirator and to perform a comparison. In addition, it was reported that there was an increase in urinary MA during the working week. This means that the excretion of MA in urine is influenced by the previous day’s exposure. We did not examine the effect of an accumulation of styrene in this study. It is necessary to compare the respirator efficiency on Monday with that on Friday to evaluate the effect of the accumulation. These are actually subjects for a future study.

There still remain many problems to solve, but workers do require to know how to wear the respirator appropriately to prevent poisoning and when should they exchange the respirator cartridge in practical situations. Therefore, it is very important to develop an easy and economical method, as precise as possible, to evaluate the efficiency of the respirator.

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