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Methods to control styrene exposure in the reinforced plastics industry

Arto Säämänen

VTT Manufacturing technology

Doctoral dissertation

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Abstract

Currently, about 75% of Finnish reinforced polyester lamination workers are exposed to styrene concentrations exceeding the current occupational exposure limit of 20 ppm. Several measures have been proposed to control styrene exposure, but only a few of them are capable of keeping the styrene concentration below 20 ppm. The present study was carried out to evaluate current ventilation systems in reinforced plastic plants and to develop new, effective ventilation systems for controlling styrene exposure. The roles of styrene-suppressed resin and styrene emission during different phases of the moulding process were also studied.

Styrene exposure and the performance of current ventilation systems were measured in 17 reinforced plastics plants. Better styrene control was sought experimentally by investigating the effects of unidirectional airflow on a worker's exposure, by testing local exhaust and local air supply ventilation methods in controlling laminator's exposure and by examining the possibilities to improve local ventilation using an automatically moving ventilation unit. The characteristics of styrene emission during hand lay-up and the effect of a rolled area on styrene evaporation were also determined.

Dilution ventilation systems are common in Finnish reinforced plastics plants. In many cases, the ventilation system was incorrectly dimensioned and workers were exposed to high styrene concentrations. High dilution airflow rates, over 850 m^3 kg^{-1}, may be needed before a concentration level below 20 ppm is achieved with the resin consumption rates common today. In order to meet the present exposure limit, local ventilation techniques should be used in combination with other control measures.

Well-designed zonal ventilation methods (e.g., a horizontal airflow tunnel) produced a favourable airflow pattern for controlling styrene vapours in the lamination area. However, work practices must be synchronized with the airflow pattern before low styrene exposure levels are reached. A worker standing in a unidirectional airflow facing downstream may also receive significant exposure due to contaminant transporting eddies formed downstream of the body. It is possible to control this type of exposure with a local air supply unit located over the worker.

Styrene control during hand lay-up moulding is also possible with the use of several local exhaust hoods. It was estimated that adequate styrene control can be achieved when the control velocity at the edge of the mould is 0.15-0.2 m s^{-1} and the exhaust airflow rate per laminate area is between 0.1 and 0.2 $m^3 s^{-1} m^{-2}$, depending on the shape of the mould. An automatically moving local ventilation unit, equipped with local exhaust and a local air supply, was able to keep the styrene concentration near or below 20 ppm when medium-size products were laminated with the hand lay-up method. A styrene concentration of below 20 ppm was also obtained when a new type of spray booth, equipped with automatically moving curtains, was used and when the control velocity was at least 0.3 m s^{-1} during the spray-up of gelcoat resin.

The use of vapour-suppressed resin reduced the total styrene emission by 30-60%, but the reduction was much lower during active lamination. After rolling, the emission from the vapour-suppressed resin was found to be very low, and, therefore, the emission rate was dependent on the size of the surface area rolled simultaneously. The use of these resins is beneficial, especially when large products are laminated.

Preface

This thesis is based on several experimental studies made at the University of Kuopio, the Finnish Institute of Occupational Health, and VTT Manufacturing Technology in 1984-1998. I wish to express my sincere gratitude to all those who have contributed to this work. The finalizing of this thesis was made possible by financial support from the Jenny and Antti Wihuri Foundation and from the Finnish Work Environment Fund, and this support is gratefully acknowledged.

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Tampere, July 1998

Arto Säämänen

Contents

Appendices of this bublication are not included in this publication. Please order the printed version to get the complete publication (http://www.inf.vtt.fi/pdf/publications/1998/)

Abbreviations and symbols

ε_a	Air change efficiency
Q	Airflow rate
ε^c	Contaminant removal effectiveness
ε_p	Local air change index
$\bar{\tau}_p$	Local mean age of air at point p
$\bar{\tau}_e^c$	Local mean age of contaminant at the exhaust
τ_n	Nominal time constant; V/Q
$\langle\bar{\tau}\rangle$	Room mean age of air (room average of local mean ages of air)
V	Room volume
ABS	Acrylonitrile-butadiene-styrene
ACGIH	American Conference of Governmental Industrial Hygienists
ANOVA	Analysis of variance
CARB	California Air Resource Board
CAS	Chemical Abstracts Service
CFA	Composites Fabrication Association
Conf. int.	Confidence interval
D.F.	Degrees of freedom
GC-FID	Gas chromatograph equipped with flame ionisation detector
GRP	Glass-fibre reinforced polyester
HSE	Health and Safety Executive

IARC	International Agency for Research on Cancer
ILO	International Labour Office
IR	Infrared
IUPAC	International Union of Pure and Applied Chemistry
LSE	Low styrene emission
MANOVA	Multivariate analysis of variance
NIOSH	National Institute of Occupational Health
NVS	Non-vapour suppressed (resin)
OSHA	Occupational Safety and Health Administration
PEL	Permissible exposure limit
ppm	parts per million (cm^3m^{-3})
REL	Recommended exposure limit
SAN	Styrene-acrylonitrile
SBR	Styrene-butadiene rubber
TLV	Threshold limit values
UP	Unsaturated polyester
VS	Vapour suppressed (resin)

List of original publications

This thesis is based on the following original publications. The thesis also contains unpublished data.

I Kalliokoski, P., Säämänen, A., Ivalo, L. & Kokotti, H. 1988. Exposure to styrene can be controlled. American Industrial Hygiene Association Journal, Vol. 49, No. 1, pp. 6-9.

II Niemelä, R., Säämänen, A., Karvinen, P., Pfäffli, P., Nylander, L. & Kalliokoski, P. 1993. Dilution ventilation to control styrene exposure in reinforced plastic industry. In: Hughes R.T., Goodfellow H.D. & Rajahns G.S. (eds.) Ventilation '91, 3rd International Symposium on Ventilation for Contaminant Control, September 16-20, 1991, Cincinnati, Ohio, USA. Cincinnati, OH, USA: American Conference of Governmental Industrial Hygienists, Inc., pp. 241 - 244.

III Säämänen, A., Niemelä, R., Blomqvist, T. & Nikander, E. 1991. Emission of styrene during the hand lay-up moulding of reinforced polyester. Applied Occupational and Environmental Hygiene, Vol. 6, No. 9, pp. 790 - 793.

IV Säämänen, A., Andersson, I.-M., Niemelä, R. & Rosén, G. 1995. Assessment of horizontal displacement flow with tracer gas pulse technique in the reinforced plastic plants. Building and Environment, Vol. 30, No. 1, pp. 135-141.

V Kulmala, I., Säämänen, A. & Enbom, S. 1996. The effect of contaminant source location on worker exposure in the near wake region. Annals of Occupational Hygiene, Vol. 40, No. 5, pp. 511-523.

VI Säämänen, A., Kulmala, I. & Enbom, S. 1998. Control of exposure caused by the contaminant source in the near wake region. Applied Occupational and Environmental Hygiene, In press.

1. Introduction

Styrene polymers are among the four major types of commercially important polymers, along with polyethylene, polypropylene and polyvinyl chloride. Therefore, styrene is one of the most important organic chemicals today. The global production of styrene was 14.3 million tonnes in 1992. The majority of the styrene was used for making various grades of polystyrene and other styrene polymer products. About 0.5 million tonnes (3%) was used in unsaturated polyester resins (European Chemical Industry Council 1993). In unsaturated polyester resin, styrene monomer is used both as a solvent and a cross-linking agent. The physical properties of the product reaches the optimum when the amount of styrene ranges from 30% to 45%, depending on the type of unsaturated polyester (Fradet & Arlaud 1989).

The commonest health problems due to styrene monomer exposure are the effects on the nervous and the respiratory systems. Symptoms include depression, concentration problems, muscle weakness, fatigue, unsteadiness, and nausea. Exposure may also irritate the nose, throat, and eyes. There is also some evidence for genotoxicity based on chromosomal changes in lymphocytes. The International Agency for Research on Cancer (IARC) has determined that styrene is a possible human carcinogen (IARC 1994). Exposure to styrene can cause adverse effects on health and well-being even in low concentrations (10-20 ppm) (Vainio 1991).

Although larger quantities of styrene are used in polystyrene, acrylonitrile-butadiene-styrene resins, styrene-butadiene resins and butadiene-styrene latex, the highest styrene exposure occurs during the handling of unsaturated polyesters, especially in the reinforced plastics industry (Pfäffli & Säämänen 1993). It has been estimated that about 2500 persons are being exposed to styrene in Finland during the manufacture of reinforced plastics products (Säämänen et al. 1991). Studies have shown that 77% of lamination workers are exposed to a styrene concentration exceeding the current Finnish occupational exposure limit of 20 ppm (Pfäffli et al. 1992). Exposure levels have slightly decreased during the last decade (Kivistö & Valkonen 1997). In Finland, exposure to styrene caused 15 registered cases of occupational disease during 1990-1996: 5 cases of nervous system disease, 4 cases of poisoning, 4 eczema and 2 cases of respiratory disease (Työterveyslaitos 1998).

Unsaturated polyesters are primarily used in reinforced plastics, usually with glass fibres. Open moulding methods, hand lay-up and spray-up are the commonest manufacturing methods used in Finland. Employees must work near large styrene-emitting surfaces with these methods, and, consequently, exposure to styrene is often heavy. Another problem, typical for small plants, is that the types, sizes and shapes of products and the number of concurrently laminated moulds change continuously. Therefore, worksites are not stationary, and it is difficult to design control measures.

Several factors affect the exposure level in reinforced plastic plants (figure 1). Successful exposure prevention can be achieved by controlling the emission of styrene during moulding and by designing the airflow field near the contaminant source and the worker carefully. Administrative control measures, as well as personal protection, can also be used to some extent.

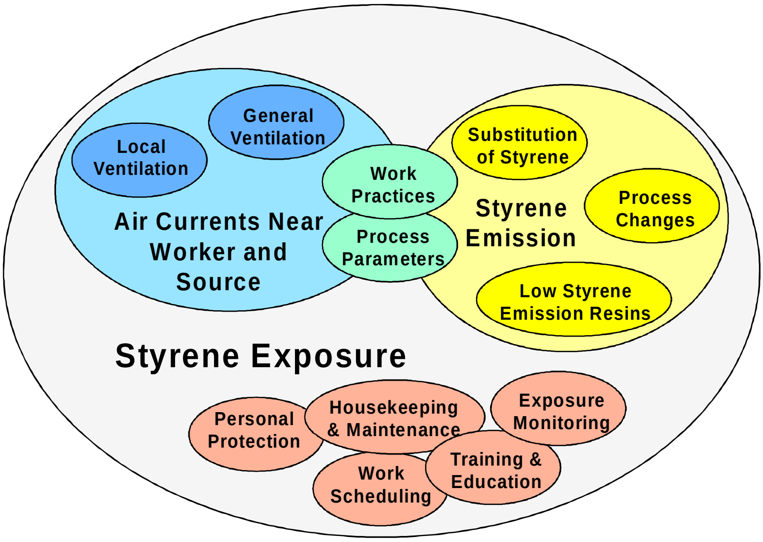


Figure 1. Factors affecting styrene exposure during the moulding of unsaturated polyesters.

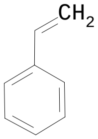
The present study was performed to evaluate the ventilation systems currently used in reinforced plastics plants and to develop new effective ventilation systems for controlling styrene exposure. The role of styrene-suppressed resin and styrene emission during different phases of the moulding process were also studied. Personal protection and administrative control measures, such as good housekeeping, training, and exposure monitoring were of minor interest in this study.

2. Review of the literature

2.1 Properties, occurrence and health effects of styrene

Styrene [CAS No. 100-42-5], also known as phenylethylene (IUPAC name) or vinylbenzene, is by far the most important industrial unsaturated aromatic monomer. Styrene is used extensively for manufacturing several plastics. It also occurs naturally in small quantities in some plants and food. Commercial processes for manufacturing styrene are mainly based on the dehydrogenation of ethylbenzene (James & Castor 1994). Styrene is a colourless liquid with a distinctive, sweetish, pungent odour. Its odour threshold is low, 0.05-0.08 ppm (Chen 1997). The most important physical properties of styrene are summarized in Table 1.

Table 1. Physical properties of styrene monomer (James & Castor 1994, Chen 1997) .

Property	**Value**
Chemical structure	
Molecular weight	104.15 g mol^{-1}
Boiling point	145.15 °C
Freezing point	-30.6 °C
Explosive limits in air	1.1-6.1%
Density	0.906 g ml^{-1}
Vapour pressure 20 °C 25 °C 50 °C 80 °C	0.6 kPa 0.87 kPa 3.2 kPa 12.2 kPa

The high reactivity of the double bond makes styrene easy to polymerize and copolymerize, even at room temperature. It is, therefore, stored in inert gas or stabilized with p-tert-butyl catechol. Styrene vapour is also very reactive in air, reacting readily with hydroxyl radicals and ozone (Alexander 1997). Styrene monomer is used to make various grades of polystyrene, acrylonitrile-butadiene-styrene terpolymer (ABS), styrene-acrylonitrile copolymer (SAN) and styrene-butadiene rubber (SBR) (Chen 1997). Minor uses of styrene include butadiene-styrene latex and unsaturated polyester resins.

Exposure to styrene can occur by inhalation and skin contact. However, the percutaneous absorption of styrene is usually an insignificant exposure route and does not contribute significantly to the body burden of workers in the reinforced plastics industry (Brooks et al. 1980). Thus occupational exposure occurs mainly via the lungs. The pulmonary uptake of styrene ranges from 60% to 70% (IARC 1994). Styrene is metabolized mainly through side chain oxidation, and it is excreted in urine as mandelic acid, phenyl glyoxylic acid and, to some extent, as hippuric acid (Pekari 1994). Main metabolites in urine, mandelic acid and phenyl glyoxylic acid, are used for the biological monitoring of styrene exposure (Aitio et al. 1997). The first metabolic oxidation product, styrene-7,8-oxide, is reactive and can form covalent binding to macromolecules (Swedish Criteria Group 1991). This reactivity may be responsible for the genotoxicity of styrene.

Styrene can have adverse effects on the health and well-being of exposed workers even at relatively low exposure levels (Forsman-Grönholm et al. 1992). Its influence on the nervous system is usually considered to be the critical effect of occupational exposure (Swedish Criteria Group 1991). Styrene is neurotoxic and impairs the central and peripheral nervous systems. Symptoms such as headache, dizziness and fatigue have commonly been observed due to exposure to styrene. Exposure may slow down reaction times and cause electroencephalographic changes, visuomotor inaccuracy and impaired co-ordination and balance (Vainio 1991, Forsman-Grönholm et al. 1992). Prolonged reaction times are common at styrene concentrations below 25 ppm (Swedish Criteria Group 1991). Increased frequencies of neuropsychiatric symptoms have been observed at exposure levels as low as 6 ppm (Swedish Criteria Group

1991). Styrene vapour has an irritating effect on the eyes and mucous membranes at high concentrations. Increased chromosomal damage has also been reported in lymphocytes (IARC 1994). However, there is inadequate evidence to show that styrene is carcinogenic in humans (IARC 1994, Kogevinas et al. 1994).

The current occupational exposure limits (8h time-weighted average) vary between 20 and 100 ppm in different countries (Table 2). The lowest exposure limits, 20-25 ppm, have been set in Nordic countries and Germany. In Finland the exposure limit was lowered from 100 ppm to 50 ppm in 1981 and further to the current 20 ppm in 1987.

Table 2. Occupational exposure limits for styrene in different countries (United States Department of Labor 1971, Arbeidtilsynet 1991, ILO 1991, Arbejdstilsynet 1992, Deutsche Forschungsgemeinschaft 1993, HSE 1993, Arbertarskyddsstyrelsen 1996, Työministeriö 1996, NIOSH 1997, ACGIH 1998).

Country/ Organization	**Exposure Limit 8 h ppm**	**Short-Term Exposure Limit ppm**
Belgium	50	100
Denmark	25	-
Finland	20	50
France	50	-
Germany	20	-
Norway	25	-
Sweden	20	50
United Kingdom	100	260
USA/ACGIH (TLV)	20	40
USA/NIOSH (REL)	50	100
USA/OSHA (PEL)	100	200

2.2 Unsaturated polyesters

Most unsaturated polyester (UP) resins are solutions of unsaturated polyesters in copolymerizable monomers which act as cross-linking agents. Several vinyl monomers can be used for this purpose. However, styrene is by far the most commonly used monomer because of its good reactivity and low cost. The physical properties of the laminate become optimal when the concentration of styrene ranges from 30% to 45%, depending on the type of UP (Fradet & Arlaud 1990). The UP ”backbone” is formed by the polycondensation reaction with unsaturated or saturated dicarboxylic acids or anhydrides and polyfunctional alcohols. UP resins are cured or cross-linked by free-radical polymerization with initiators, usually benzoyl peroxide or methyl ethyl ketone peroxide. These organic peroxides are dissociated by heat or redox metal activators into peroxy and hydroperoxy free radicals. The free radicals initially formed are neutralized by quinone stabilizers, which temporarily delay the cross-linking reaction. This temporary induction period between the addition of the initiator and the change in the semisolid gelatinous mass is referred to as gelation time (gel time). As the stabilizer is consumed, the peroxy radicals initiate the addition chain propagation reactions between the unsaturated fumarate groups of polyester chains and the double bonds of the solvent monomer (Krämer 1992, Chen 1997).

The majority of UP resins are reinforced with glass fibres, which impart high strength and rigidity to fabricated parts. Reinforced plastics products can be fabricated using several processes, depending on the desired physical characteristics. The principal processes include hand lay-up, spray-up, continuous lamination, filament winding, pultrusion and various closed moulding operations. The simplest and most commonly used fabrication method is hand lay-up moulding. It is a manual technique employing open moulds; the reinforcement mat or woven fabric is manually fitted to a mould and saturated with initiator-resin mix. Layers of reinforcement and resin are added to build the desired laminate thickness. Brushes and rollers are used to compact each layer as it is applied. In many cases, the mould is first sprayed with gel coat, a pigmented resin mix that forms the smooth, durable outer surface of the product. Spray-up moulding uses mechanical spraying and reinforcement filament chopping equipment for depositing the resin and glass reinforcement simultaneously on the mould. After the spraying, the laminate is consolidated with tools similar to those used in hand lay-up moulding.

2.3 Exposure in reinforced plastics plants

A considerable amount of styrene monomer evaporates during the fabrication of products made of glass-fibre reinforced polyester (GRP). Therefore, styrene is the major air contaminant in this industry, and the workers' exposure level may become heavy. Mean exposures during lamination work commonly exceed 20 ppm. The means have varied between 17 and 110 ppm (Crandall 1981, Schumacher et al. 1981, Guillemin et al. 1982, Ikeda et al. 1982, Lemasters et al. 1985, Sullivan & Sullivan 1986, Jensen et al. 1990, Bellander 1992, Geuskens et al. 1992, Pfäffli et al. 1992, Sala & Catelani 1992, Sass-Kortsak et al. 1996). Although exposure has decreased during the last few decades, only fewer than 20% of lamination workers have mean styrene exposure levels lower than 20 ppm (Jensen et al. 1990, Pfäffli & Säämänen 1993). According to the biological monitoring measurements, exposure has decreased only slightly during the last decade in Finland (Kivistö & Valkonen 1997).

Exposure to styrene is high, especially when open moulding methods are used. Chopper gun operations, hand lay-up moulding and filament winding are the methods producing the highest airborne styrene concentrations, with geometric means ranging from 42-130 ppm, to 13-101 ppm and 19-125 ppm, respectively (Crandall & Hartle 1985, Sullivan & Sullivan 1986, Geuskens et al. 1992, Pfäffli et al. 1992, Sass-Kortsak et al. 1996). In these operations, high short-term exposure peaks up to 580 ppm have been observed (Geuskens et al. 1992). Exposure during gel coating is lower since these jobs are generally performed in paint booths and the application rate of the resin is commonly low (Schumacher et al. 1981, Sullivan & Sullivan 1986). On the other hand, the styrene concentration during closed moulding is normally below 20 ppm (Pfäffli et al. 1992, Sass-Kortsak et al. 1996).

Workers participating in boat-building operations have greater exposure to styrene than laminators in other types of industries (Kalliokoski 1976, Schumacher et al. 1981, Sullivan & Sullivan 1986, Säämänen et al. 1991, Pfäffli et al. 1992). Especially, the lamination of hulls and decks has produced high exposure levels (Schumacher et al. 1981). However, the exposure may be over 20 ppm also during the lamination of small parts (Crandall 1985, Pfäffli et al. 1992).

The use of vapour-suppressed resins has reduced styrene exposure by 27-50% when compared with the use of standard resin (Kalliokoski et al. 1984, Pfäffli et al. 1992, Andersson et al. 1994). In addition, 56% lower styrene concentrations in general workplace air have been detected in the plants using vapour-suppressed resin (Pfäffli et al. 1992).

Other factors affecting styrene exposure are the distance of the worker from the surface from which styrene is evaporating (Sullivan & Sullivan 1986) and the amount of polyester resin handled (Geuskens et al. 1992). Occupational exposure depends also to some degree on the area of other wet surfaces (spillage, wet tools, open buckets, etc.) in the vicinity of the worker (Andersson et al. 1994). In addition, the use of long handled tools reduce the exposure (Pfäffli et al. 1992). It is typical for the Finnish GRP industry that exposure to styrene has been heavy in small (<5 workers) and medium-size (5-20 workers) plants. Pfäffli et al. (1992) found that the mean exposures in these plants were 110 ppm and 46 ppm for laminators and 53 ppm and 14 ppm for other workers, respectively. In large plants (>20 workers), the mean exposure to styrene was lower, although it also exceeded the 20 ppm level.

Other chemicals are also used in the GRP industry (e.g., initiators for polymerization and various solvents in waxes, in other releasing agents, and in various cleaning operations). The most frequent co-contaminant for styrene is acetone, which is mainly used for cleaning tools. However, the risk of exceeding the exposure limit of acetone is much lower than that for styrene (Jensen et al. 1990, Pfäffli et al. 1992). Other solvents, styrene-7,8-oxide and methylethylketone peroxides are detected in workshop air at low concentrations (Fjeldstad et al. 1979, Pfäffli et al. 1979, Purnell et al. 1979, Jensen et al. 1990., Pfäffli 1992, Pfäffli et al. 1992, Pfäffli &

Säämänen 1993). A rough estimation of a ratio of styrene-7,8-oxide and styrene concentrations is 1:1000 (Pfäffli & Säämänen 1993). The dust from cutting, grinding and drilling may also cause hygienic problems in GRP plants (Antonsson & Runmark 1987, Pfäffli et al. 1992). Dust exposures occur mainly during assembly and finishing operations.

2.4 Exposure control technologies

Several strategies can be used to reduce styrene exposure. The commonest controls applied at the emission source include material substitution, process or equipment modifications, isolation, local ventilation, and good work practices and housekeeping. Several studies have been done to find efficient control measures for styrene exposure in the GRP industry. Most of the work has been done to develop efficient ventilation systems and to reduce styrene evaporation from the resin.

2.4.1 Resin technology

The styrene monomer reacts with the unsaturated polyester in a cross-linking reaction, and a rigid network is formed. Although the styrene monomer is aimed at becoming a part of the polymer network, some of it evaporates into the work environment before the polymerization is complete. The exothermic nature of the polymerization reaction also contributes to the evaporation. The temperature inside the laminate is commonly 40-45°C (maximum 50-60°C) during curing, whereupon the vapour pressure of the styrene is increased markedly. However, an increase in temperature occurs mainly after the gel time when the lamination work has been finished.

Several efforts have been made to minimize the evaporation of styrene during moulding. The three main approaches involve substitution of styrene with low vapour pressure monomers, high-solid unsaturated polyesters with reduced styrene content, and vapour-suppressed resins which contain styrene emission inhibiting additives.

The substitution of styrene with another, less volatile monomer would be an efficient way to reduce the emission. Any vinyl, allyl or acrylic compound fulfils the general requirements for free radical copolymerization and can be used in place of styrene (Krämer 1992). Several monomers have been proposed as alternatives. The most promising are vinyltoluene, p-methylstyrene, and low molecular weight acrylate and methacrylate monomers (Skrifvars 1995). Tests done with p-methylstyrene and vinyltoluene, which have 4 times lower vapour pressures than styrene (0.15 kPa at 20°C, Chen 1997), showed that monomer emission behaves as expected and a reduction of 70%, when compared with the emission of styrene-containing resin, was obtained during rolling (Skrifvars et al. 1992). Emission reductions achievable if low vapour pressure monomers are used depends on the monomer and the amount of styrene replaced. However, these alternative monomers are more expensive than styrene and of less commercial availability. In addition, the health effects of neither of these monomers have been investigated as much as those of styrene .

The styrene content of conventional resin is typically around 41-45%. High-solid resins contain typically less than 35% styrene, but resins with a styrene content down to 20% have been introduced (Skrifvars 1995). The use of low styrene resin (35% styrene versus 43% styrene) can potentially reduce the total styrene emission by 14-19% (Stockton & Kuo 1990, Radian Corporation 1993). Lacovara et al. (1996a) have also shown that a decrease from 42% to 35% in styrene content yields an average decrease in emission of 16% per weight of available styrene during hand lay-up, 10% during gel coating and 35% during spray-up. The overall reduction in employee exposure has been 30% to 50% (Walewski & Stockton 1985). Low styrene resins are more viscous than conventional resins and therefore make the processing difficult and reduce the applicability of the resins, especially for products with strict strength requirements (Stockton & Kuo 1990).

Vapour-suppressed resins contain additives that reduce styrene emission. Paraffin or wax compounds are typical additives that form a film on the laminate surface and reduce the evaporation of free styrene during resin curing. According to static gravimetric evaporation tests, the use of these agents decreases emission levels by 50-90% (Duffy 1979, Heilmann & Cope 1985). However, emission reductions have been lower in field studies, ranging from 30% to 70% (Stockton & Kuo 1990, Andersson et al. 1994). One drawback of these additives has been their deleterious effect on the secondary bonding of new laminate layers (Stockton & Kuo 1990). Surface sanding or solvent wiping has been necessary before the next layer has been laminated. However, newer reports show that problems with interlaminar adhesion have been solved and that styrene-suppressed resins can be used without time-consuming sanding or solvent wiping (Skrifvars 1995).

2.4.2 Emission of styrene

The amount of styrene evaporating during the processing of UP resins depends primarily on the application process, the type of UP resin, the type of product, and the ambient conditions. The highest amount of styrene evaporates during the application of the gel coat (Table 3). In many cases, gel coat is applied to the mould by spraying, which produces a high styrene emission rate. During spray-up, a large percentage of styrene is also evaporated. This evaporation, combined with the large application rate of resin, produces a high styrene emission rate. Hand lay-up moulding is the commonest application method in Finland. The emission rate is moderate during this operation, especially when vapour-suppressed resins are used. The rolling of the laminate after spraying is comparable to hand lay-up. The emission of styrene during this phase is important because the rolling phase accounts for 20% to 50% of the total exposure (Andersson & Rosén 1994, Andersson & Rosén 1995). In general, closed moulding methods produce low styrene emissions.

Several other factors influence styrene emission during moulding. An increase in the temperature of the resin or the workroom air increases the emission (Radian Corporation 1993). A large surface area or a thin laminate allows much evaporation in terms of total mass (Radian Corporation 1993, Lacovara et al. 1996a). A long gel time increases the styrene emission when conventional or high-solid resins are used, but it has no effect with vapour-suppressed resins (Milleville & Swiech 1993, Lacovara et al. 1996a). On the other hand, the airflow over the mould surface is an insignificant factor, contrary to what is commonly believed (Ivalo et al. 1985, Lacovara et al. 1996a). In addition, the spray gun set up and the spraying technique have been shown to influence styrene emission. A large spray fan angle, a high spraying pressure and a long spraying distance increases the evaporation of styrene, while a large diameter of the nozzle decreases the emission (Lacovara et al. 1996b, Säämänen & Skrifvars 1996).

Table 3. Comparison of styrene emission factors for different operations (Radian Corporation 1993, Kalliokoski et al. 1994, Lacovara et al. 1996a)

Process	**Resin type**	**Total styrene emission (% of available styrene)**		
		CARB	**CFA**	**Kalliokoski et. al 1994**
Hand lay-up	NVS	16-35	10-21	12
	VS	14-20	/	4
	Gel coat	47	/	/
Spray-up	NVS	9-13	17-38	/
	VS	5-9	/	14
	Gel coat	16-35	44-62	27
Hand and spray	NVS	11-19	/	/
	VS	6-13	/	8
	Gel coat	31-38	/	/
Filament winding	NVS	6-13	/	10
	VS	3-9	/	/
Closed moulding	NVS	1-3	/	3
	VS	1-3	/	/

2.4.3 General ventilation

Ventilation is one of the most important engineering control measures. It is also important from the viewpoint of thermal climate and energy consumption, particularly in the Nordic countries. Ventilation methods are normally classified roughly into dilution and local ventilation. However, in this study ventilation methods, which are not purely dilution or local ventilation methods and are producing localized well-ventilated zones within the workroom, are called zonal ventilation.

Dilution ventilation is widely used to control styrene vapours in GRP plants. The advantage of dilution ventilation is that it permits free mould placement within the workshop. Its disadvantage is the requirement for large volumes of fresh air, thus causing high heating costs in winter. In addition, the distance between the worker and the large surfaces from which styrene evaporates is also so small and the emission rate so strong that dilution ventilation is ineffective, resulting in high exposure to styrene. In many cases the mixing of the air remains incomplete and an increased dilution airflow rate reduces the local styrene concentrations only marginally (Sullivan & Sullivan 1986).

Well-ventilated zones in lamination rooms utilizing controlled airflows over the moulds also have be used to control styrene exposure during hand lay-up moulding. The use of auxiliary fans has been effective in creating localized airflow and removing styrene vapours from the worker’s breathing zone (Kalliokoski 1976, Sullivan & Sullivan 1986). However, this approach may disseminate vapours throughout the entire workroom and expose all the workers to low levels of styrene (Sullivan & Sullivan 1986). In the push-pull system, a fan is designated to push uncontaminated room air into the concave mould and then be exhausted at the other end into a hood (Videm 1979, Todd 1985). This system controlls the average styrene exposure level to 28 ppm during the lamination of a 14 m (46 ft) boat hull (Todd 1985).

Good results have been obtained in tunnel-like lamination rooms where a horizontal airflow pattern is produced by supplying fresh air through a low-impulse inlet at one end of the room and exhausting it from the opposite end. Using the GridMap technique, Andersson et al. (1993a) showed that, in this kind of ventilation system, styrene vapour is carried by the airflow from the wet surface toward the outlet. During hand lay-up moulding the laminator's mean exposure ranges from 5 to 11 ppm (Andersson et al. 1993a).

Systems utilizing vertical local air supply over the lamination zone are also used to reduce styrene exposure. This kind of ”air shower” was found to reduce styrene exposure by 73-100% during the lamination of small and medium-size products (Andersson & Rosén 1993, Andersson et al. 1993c, Andersson 1995). With this system, exposure levels below 10 ppm were achieved during the lamination of bath tubes (Andersson et al. 1993c). However, in operations extending outside the reach of the system, the performance of the local air supply was diminished (Andersson et al. 1993c, Andersson & Rosén 1993).

2.4.4 Local ventilation

Local exhaust ventilation extracts vapours close to the source. It is inherently more effective than dilution ventilation. It is possible to control styrene exposure to very low levels when a large volume of air is exhausted with a large hood or several hoods. Isaksson (1976) obtained a styrene concentration below 20 ppm when 0.56 $m^3 s^{-1}$ to 1.66 $m^3 s^{-1}$ of air was exhausted through 1 to 4 hoods during the lamination of a 1.6 m^2 plate and 3 m rowboat. Todd (1985) used an exhaust trench on the floor below the tilted 12.5 m (41 ft) and 14 m (46 ft) boat hull. The exhaust flow rate through 5 exhaust slots was 42 $m^3 s^{-1}$, and the average exposure became 21 ppm. On the other hand, Ikeda et al. (1982) reported that the styrene concentration was as high as 65 ppm when the exhaust flow rate was 3 $m^3 s^{-1}$ from a 14.5 m boat hull through 3 exhaust hoods. Exhaust hoods have the disadvantage that they are difficult to adjust and move around the mould and their suction area is small. Therefore, adequate control may not be achieved, especially when large moulds are laminated. The use of exhaust ducts is rather uncommon in GRP plants (Sullivan & Sullivan 1986).

Local exhaust ventilation can operate effectively when the area of the contaminant source is small and the work process is stationary. In recent years, systems combining local exhaust ventilation and local supply ventilation have been introduced for various work tasks (Volkwein et al. 1988, Gressel & Fischbach 1989, Chamberlin 1990, Eloranta et al. 1993, Heinonen et al. 1996). Such local ventilation systems have also been used successfully in the reinforced plastics industry. One such system is the ”local ventilation unit”, which consists of a local exhaust slot and an air supply unit (Andersson 1995). With such a unit, the worker’s exposure is reduced as much as 88% when small products ($<0.5\ m^2$) are laminated (Andersson et al. 1993b). In the lamination of somewhat larger products, a reduction of 60% has been attained (Andersson et al. 1993b).

Spray-up operations, especially gel coat spraying, take place mainly in spray booths (Crandall 1981, Schumacher et al. 1981, Sullivan & Sullivan 1986). Booths with a horizontal airflow, called side-draught booths, are normally used in GRP plants. This is the commonest flow direction in spray booths, and it takes advantage of the momentum of the spray (O’Brien & Hurley 1982). Control velocities from 0.3 to $1.5\ m\ s^{-1}$ are recommended for spray booths (Fletcher 1989, ACGIH 1992). However, such velocities normally lead to very high airflow rates through the booth. One possible way to reduce the airflow rate of the booth is to reduce the area of the open face, for example, with curtains (Isaksson 1976). However, the movement of the curtains may be cumbersome as the work is passed on. Automatically moving curtains have been used to solve this problem (Andersson & Rosén 1994). It has been possible to reduce the exhaust airflow rate from $11.1\ m^3\ s^{-1}$ in the ordinary spray booth to $2.1\ m^3\ s^{-1}$ in the ”IVF-booth” with moving frames during the spray up lamination of a large boat.

Most of the previously presented zonal and local control technologies utilize horizontal or vertical unidirectional airflow as a control measure. However, the utilization of such an airflow may cause some problems. Several authors have shown that a person in a unidirectional flow may generate a wake region downstream of the body (Ljungqvist 1979, Flynn & Shelton 1990, Geroge et al. 1990, Kim & Flynn 1991a, Flynn & Ljungqvist 1995, Johnson et al. 1996). If the contaminant source and the worker’s breathing zone is within this wake region high exposure may occur.

2.4.5 Other control measures

Changing the process from open contact moulding to closed moulding should decrease styrene exposure dramatically. However, open moulding is more commonly used because of the numerous types and small series of products fabricated. Continuously changing production usually makes the closed moulding methods too expensive. On the other hand, the development of conventional lamination processes may also lead to reduced exposure. The use of spray guns with high transfer efficiency and the optimization of spraying parameters have been shown to decrease styrene emission (Lacovara et al. 1996b). The use of long handled tools increases the distance between the surface of the laminate and the worker and results in lower exposure (Kalliokoski et al. 1984). The passive exposure of non-laminators can be reduced by isolating them from the lamination area.

Because inhalation is the major route of entry into the body for styrene, respiratory protection is, in principle, an effective control method (Brooks et al. 1980, Ikeda et al. 1982, Löf et al. 1993, Kawai et al. 1994). The use of respirators has yielded reductions of 30-99% in styrene exposure measured with biological monitoring methods (Ikeda et al. 1982, Löf et al. 1993, Kawai et al. 1994). This wide range may be due to the fact that the workers used respirators only a part of the work period (Löf et al. 1993). However, respirators are uncomfortable to use and only few subjects are reported to use them in GRP plants (Sullivan & Sullivan 1986, Sass-Kortsak et al. 1996). Air-purifying respirators are not recommended for continuous use due to their high breathing resistance (Louhevaara 1985).

Training workers in correct work practices and housekeeping has been shown to reduce styrene exposure markedly (Hopkins et al. 1986). It was noted that engineering controls were not very effective until behavioural controls were introduced (Hopkins et al. 1986). Proper work behaviour includes keeping breathing zones away from sources of styrene and working upwind of the sources, as well as enforcing engineering controls

(Hopkins et al. 1986, Andersson et al. 1993a). In order to maintain good work practices, continuing feedback may be needed as part of reliable behavioural control technology (Hopkins et al. 1986). It is obvious that resin modifications and the implementation of user friendly local ventilation are the key control measures for styrene during GRP manufacturing. In addition, the careful training and motivation of workers are necessary to obtain effective exposure control (Hopkins et al. 1986, Andersson 1995).

2.5 Current status of control technologies

During the past decade, the exposure limits for styrene have been lowered in many countries, especially in the Nordic countries. However, control technology has not followed this development fast enough. The use of low styrene content or vapour-suppressed resins has helped to reduce styrene emission, but it has not solved the exposure problem (Kalliokoski et al. 1984, Pfäffli et al. 1992, Andersson et al. 1994). Heavy exposure to styrene remains a common problem in the GRP industry, and most of the workers are exposed to styrene concentrations exceeding the current Finnish exposure limit of 20 ppm. Therefore new control innovations are badly needed for GRP moulding work.

The types, sizes and shapes of produced items and the number of concurrently laminated moulds are changing continuously and therefore workstations cannot be stationary. This is also the reason why dilution ventilation is often used in GRP plants.

The utilization of localized airflows over the mould with the aid of auxiliary fans is one of the most often used control measures (Kalliokoski 1976, Videm 1979, Todd 1985, Sullivan & Sullivan 1986). However, it may be possible to create an airflow pattern similar to that of auxiliary fans with the aid of high-velocity recirculating air jets. A horizontal airflow pattern in a tunnel-like lamination room has also been used with good results to control styrene vapours (Andersson et al. 1993a).

Local exhaust ventilation has been successfully used in GRP plants. However, large airflow rates and a great number of exhaust openings are needed to keep exposure below the 20 ppm limit. However, it may be possible to control styrene vapours with exhaust hoods using a smaller exhaust airflow rate and a sufficient number of exhaust hoods if the ventilation system moves automatically. Another means for improving the performance of local exhaust ventilation has been the use of local supply air together with exhausts. This method has been studied during the lamination of small objects (Andersson et al. 1993b). It may also be possible to improve worker protection in GRP plants by using a combination of exhaust ducts and a local air supply during the lamination of medium-size objects.

Spray-up lamination is normally performed in spray booths. In these applications, high exhaust airflow rates are needed. It may be possible to reduce the worker’s exposure and the exhaust airflow rate of the booth, at the same time, with the combination of moving curtains and a local air supply over the worker.

Many of the previously presented control measures utilize a unidirectional airflow pattern near the worker to keep styrene vapours away from the breathing zone. However, several authors have pointed out that this kind of airflow pattern can produce a wake region downstream of the body and thus impair the protection of the control measure. In most of the studies, the contaminant source has been fixed or the distance varied at one height only (Ljungqvist 1979, George et al. 1990, Kim & Flynn 1991b). Thus there is insufficient knowledge on the 3-dimensional transport phenomenon within this wake region, and only little information is available on the control of the wake region.

3. Aims of the study

The occupational exposure limit for styrene is often exceeded in GRP plants. The purpose of this study was to develop and evaluate efficient control measures for styrene vapours in GRP plants. The detailed objectives of this work were:

- to evaluate the efficiency of current ventilation systems in Finnish GRP plants (Paper III)

- to define the characteristics of styrene emission by

    - investigating the evaporation rate during various phases of moulding and comparing styrene evaporation from standard and styrene suppressed (low styrene emission, LSE) resins (Paper II)

    - studying the effect of the area being rolled on styrene evaporation (Section 5.1.2)

- to improve the ventilation methods used in GRP plants by

    - investigating the use of recirculating air jets and a horizontal flow tunnel in creating a unidirectional airflow pattern for lamination areas (Paper I, IV)

    - examining the formation of the wake region downstream of the worker in unidirectional airflow and controlling the contaminant transport from this area to the worker’s breathing zone (Paper V, VI)

    - studying the use of local exhaust ducts (Paper I) and automatically moving local ventilation units during hand lay-up (Section 5.3.3) and spray-up moulding (Section 5.3.4)

4. Materials and methods

This research consisted of several studies in which different parameters and contaminants were studied using various sampling devices. A concise description of these parameters and contaminants are presented in table .

Table 4. Summary of the study methods used in this project.

Short study name	**Study place**	**Measured parameter**	**Contaminant**	**Sampling device**	**Reference**
Styrene emission during hand lay-up	Laboratory	Emission	Styrene	IR analyser	Paper III
Effect of rolling area on emission	Laboratory	Emission	Styrene	IR analyser	Section 5.1.2
Ventilation in Finnish GRP plants	Field study	Exposure	Styrene	Passive dosimeter	Paper II
Recirculation air jets	Field study	Exposure	Styrene	Charcoal tube	Paper I
Horizontal flow tunnel	Field study	Ventilation efficiency	Tracer gas	IR analyser	Paper IV
Local exhaust with large moulds	Field study	Exposure	Styrene	Charcoal tube	Paper I
Formation and control of wake	Laboratory	Contaminant transport	Tracer gas	IR analyser	Papers V, VI
Comparison of ventilation systems	Laboratory	Exposure	Styrene	Charcoal tube	Section 5.3.3
Improvement of spray-up booth	Laboratory	Exposure	Styrene	Charcoal tube	Section 5.3.4

The characteristics of styrene emission during hand lay-up, as well as the effect of the area being continuously rolled on styrene evaporation, were investigated under laboratory conditions. In these studies, the concentration of styrene was monitored in the exhaust duct of the study chamber with an infrared (IR) analyser. Ventilation studies were conducted both in the laboratory and in 22 GRP plants. The efficiency of various ventilation arrangements was compared by measuring the concentration of styrene in the worker's breathing zone. A tracer gas (nitrous oxide or sulphur hexafluoride) was used to investigate both airflow patterns in a horizontal airflow tunnel and the recirculation of contaminated air near the worker's body.

4.1 Measurement of styrene concentration

When the performance of current ventilation systems in Finnish GRP plants was studied, the time-weighted average full-shift styrene concentration (8h) in the laminator's breathing zone was determined using passive dosimeters. Simultaneous full-shift styrene sampling was also carried out at stationary sampling points.

When different ventilation methods were compared, styrene concentrations were measured only during the active lamination phases so that comparable styrene concentrations would be available. Charcoal tube sampling was used for both the personal and area sampling. The variation of styrene concentration was followed with the infrared (IR) analyser at some stationary sampling sites and in the exhaust duct.

In the study of current ventilation systems in Finnish GRP plants (paper II), the average full-shift styrene concentrations (8h) in the laminators' breathing zone were measured using passive dosimeters (3M No. 3500 - Organic Vapor Monitor, 3M, St. Paul, MN, USA). Two samples per shift were taken. Styrene samples were also collected at area sampling points (height 1.5 m) in 1-4 locations within the lamination area. For this purpose, activated charcoal sampling tubes (SKC 226-01, 20/40 mesh, SKC Inc. Eighty Four, PA, USA) were used together with personal sampling pumps (SKC 222-3, SKC Inc. Eighty Four, PA, USA) operating at an airflow rate of 100 ml min^{-1}. Passive dosimeters and charcoal tubes were desorbed with 2 ml of mixture containing 30% carbon disulphide and 70% dichloromethane. The samples were analysed with a gas chromatograhph equipped with a flame ionization detector (GC-FID). A more-detailed description of the analysis of the styrene samples has been reported elsewhere (Pfäffli et al. 1992).

The styrene sampling time was shorter in the other ventilation studies (Paper I and Sections 5.3.3 and 5.3.4). Activated charcoal tubes and personal pumps were used. The samples (paper I) were desorbed using carbon disulphide or dichloromethane and analysed with GC-FID (Kalliokoski & Pfäffli 1975). In the more recent studies (Sections 5.3.3 and 5.3.4), charcoal tubes were analysed as in Paper II (Pfäffli et al. 1992).

The variation of the styrene concentration within the workroom areas and in the exhaust air was monitored using an IR analyser (Miran 1A, Foxboro Inc., South Norwalk, CT, USA). The wavelength was 11.1 µm and the path length 21.75 m. The analyser was calibrated using the closed loop calibration method (Wilks 1975). The signal was recorded with a strip chart recorder (paper I) or stored in a computer (papers II and III and sections 5.1.2 and 5.3.4). The sampling time was 30-60 seconds, and the sampling airflow rate was 20 l min^{-1}.

4.2 Determination of ventilation efficiency parameters

Air and contaminant movements in the lamination room were investigated with the tracer gas technique using either step-down (Paper II) or pulse releasing methods (Paper IV). The results of the tracer gas experiments were interpreted by the age analysis. From the age values, the air change efficiency, local air change index and the contaminant removal effectiveness were calculated (Sutcliffe 1990, Brouns & Waters 1991).

Nitrous oxide (N_2O) was used as the tracer gas. The injection equipment consisted of a nitrous oxide tank with a pressure-reducing valve, a calibrated rotameter and a dry gas volume meter. The concentration of nitrous oxide was continuously monitored with an IR analyser (Miran 1A, Foxboro Inc., South Norwalk, CT, USA). The wavelength used was 4.45 µm and the path length was 20.25 m. The tracer gas releasing and measuring points and formulas used to determine the efficiency parameters are presented in table 5.

Table 5. Tracer gas injection and monitoring points and the formulas used to determine the different ventilation efficiency parameters.

Parameter	Formula used	Injection point	Measuring point	Paper
Air change efficiency	$\varepsilon_a = \frac{\tau_n}{2\langle\bar{\tau}\rangle}$	Supply air	Exhaust air	II, IV
Local air change index	$\varepsilon_p = \frac{\tau_n}{\bar{\tau}_p}$	Supply air	Point in occupied zone	IV
Contaminant removal effectiveness	$\varepsilon^c = \frac{\tau_n}{\bar{\tau}_e^c}$	Point within lamination room	Exhaust air	IV

4.3 Measurement of styrene emission

4.3.1 Emission during hand lay-up

The characteristics of the styrene emission during lamination work were studied both in the laboratory and in the field. Styrene emission was determined by measuring the exhaust airflow rate and the styrene concentration in exhaust air. The styrene measurements were made with the IR analyser as has already been described. The styrene emission rate was then obtained by multiplying the flow rate by the concentration.

The styrene emission rates were measured for two polyester resins (paper III). The first resin was LSE type containing vapour-suppressant additives, whereas the second was a conventional (standard) resin without additives.

Both were commercially available orthophthalic resins. Test laminates (0.165 m^2) were moulded in a small ventilated enclosure built inside the laboratory hood (Figure 1, Paper III). The temperature inside the laminate was also followed with T-type thermocouples.

The emission of styrene was also measured in one field study (Paper II). In this study, the concentration of styrene was monitored in the exhaust duct until the concentration decreased to below 1 ppm. Only the total mass of styrene evaporated was measured during the work shift. The total styrene emission was obtained by monitoring the total dose of styrene in the exhaust duct and multiplying it by the flow rate.

4.3.2 Effect of the rolled area

The effect of a rolling area on styrene emission was studied in the laboratory during the roll out of sprayed laminate (Section 5.1.2). The measurements were conducted in a measurement tunnel, which was 5 m long, 2 m wide and 2 m high (figure). Air flowed freely into the tunnel from the surrounding laboratory hall. The air was exhausted from the other end of the tunnel into a duct. The airflow rate through the tunnel was 0.9 $m^3 s^{-1}$, and it was measured using a calibrated orifice. The concentration of styrene was determined with the IR analyser in the exhaust duct.

The mould (area 3 m^2) used in this study was a plane inclined 45°. Firstly, a known amount of LSE resin (M105TB, Neste Polyester, Finland) and chopped glass fibre (2400 tex, Owens Coring, Great Britain) was sprayed on the mould. The quantities planed were 10 000 g of resin and 4 300 g of glass fibre to give a final glass-to-resin ratio of 1:3. After spraying, the styrene vapours due to the spray-up were ventilated out of the tunnel. The mould was left untouched inside the tunnel for 5 minutes. It was then rolled out using the designed rolling pattern, and the emission of styrene was determined.

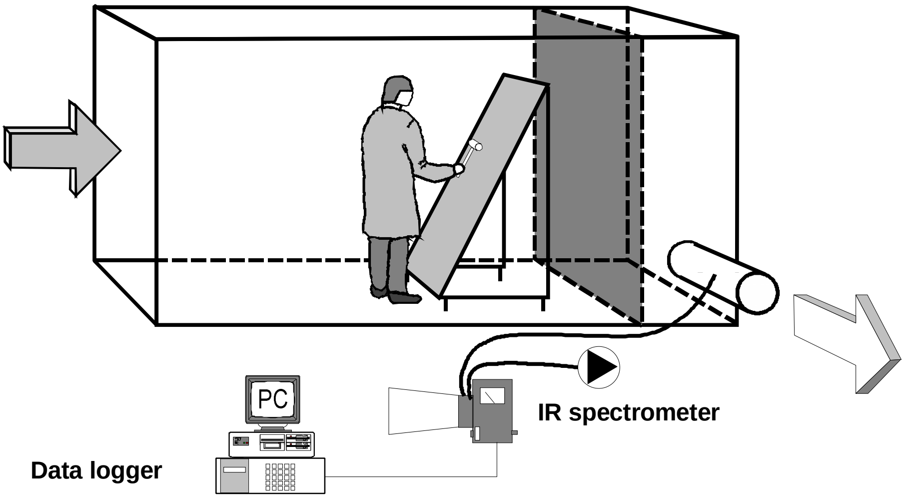


Figure 2. Measurement tunnel used in the rolling area experiments.

The continuously rolled areas were 1/1, 1/2 and 1/4 of the mould area (3 m^2, 1.5 m^2 and 0.75 m^2, respectively). The time spent rolling out the entire mould was kept constant (5 minutes). Therefore the entire mould area was rolled 5 minutes in the 1/1 test. In the 1/2 test, half of the mould (1.5 m^2) was rolled within 2.5 minutes, after which the second half of the mould was rolled for another 2.5 minutes. In the 1/4 test, each quarter of the mould was rolled for 1.25 minutes. Each test was repeated 7 times in random order. The concentration of styrene was determined in the exhaust duct with the IR spectrometer. The styrene emission rate (mg s^{-1}) during the spraying was calculated from the ”steady state” styrene concentration in the duct. The measurement was started 60 seconds after the spraying began, and it was stopped when the spraying ended to avoid the dilution effect of the ventilation system. It was assumed, based on the tunnel volume and airflow rate, that the steady state was achieved within 60 seconds. The styrene emission rate was obtained by multiplying the average concentration by the airflow rate. The results were analysed using a oneway analysis of variance (procedure ONEWAY, SPSS for Windows, Release 6.1.3, SPSS Inc., USA), and the comparison of the group mean values was done using Duncan’s multiple range test (Montgomery 1991).

4.4 General ventilation studies

4.4.1 Evaluation of current ventilation systems

Field measurements were made in 17 Finnish reinforced plastics plants, 15 of which used the open moulding process and two of which employed closed moulding techniques (Paper II). Most of the plants manufactured boats, but vehicle parts, tubes and silos, as well as sandwich panels, were also produced. Hand lay-up moulding was the commonest production method. Filament winding, chopper gun operations and gel coat spraying were also used. These sites were a part of a larger Finnish GRP investigation in which styrene exposure measurements were done in 32 randomly chosen plants (Pfäffli et al. 1992).

All the plants used mechanical supply and exhaust ventilation. The fresh air was generally supplied at a relatively high velocity, and it resulted in effective mixing of the fresh air with the room air. Systems in which the main airflow was controlled by means of horizontally and vertically angled high-momentum air jets were used in four rooms. The exhaust hoods were generally wall mounted, but flexible and movable local exhaust ducts were installed in five plants. However, they were not located properly (i.e., they were not close to the styrene sources), and, therefore, they functioned mainly as exhausts for general ventilation.

The styrene exposure of 105 laminators working in the 17 plants was investigated. Styrene concentrations in general lamination room air were also measured at stationary sampling points. The supply and exhaust airflow rates were determined using the Pitot-tube traversing technique. The styrene concentration in the exhaust air and the exhaust airflows were used to calculate the styrene emission from the plant. Records on resin type, styrene content of the resin and the amounts of resin consumed were kept in each plant. The air change efficiency was determined with tracer gas using the step-down method.

4.4.2 Zonal unidirectional airflow systems

The effects of zonal unidirectional airflow on workers' styrene exposure and on airflow patterns were investigated in four GRP plants. Two of them utilized recirculation air jets (Paper I). In two other plants, unidirectional airflow was produced in a tunnel-like lamination room (Paper IV).

In both plants utilizing recirculation air jets, make-up air supply and exhaust openings were located on opposite walls (Figures 1 and 2, Paper I). The effect of mould position in the plant and in relation to the airflow and the effect of the recirculation air jets on the styrene concentration in the laminators' breathing zone were studied. Concentration fluctuation was monitored with an IR analyser at the stationary sampling points. Air velocities near the mould were measured with a thermoanemometer (GGA-23S, Wallac Oy, Finland). Smoke tests were also performed to demonstrate airflow near the moulds.

In tunnel-like lamination rooms, the supply air was introduced through a large perforated plenum and was exhausted through a large air outlet on the opposite wall (Figure 1, Paper IV). The performance of this ventilation system was evaluated in a plant where large boats, up to 17 m long, were manufactured. The tracer gas pulse technique was used to measure the air change efficiency, local air change index, and the contaminant removal effectiveness.

4.5 Local ventilation studies

4.5.1 Wake effects near the worker

The experiments on wake effects were carried out in an open-ended tunnel, 2 m high × 2 m wide × 4.2 m deep (Paper V, VI). The air flowed freely into the tunnel from the surrounding laboratory hall and was exhausted through a perforated plenum so that the air distribution would be even across the tunnel (Figure 1, Paper V). An anthropometric mannequin, 1.52 m tall and 0.38 m wide at the shoulder, was facing downstream at the centreline of the tunnel. The transport of the contaminants from the source within the near wake region to the worker's breathing zone was examined by releasing tracer gas from 420 points downstream of the mannequin and measuring the tracer concentration in the middle of the mannequin's nose and mouth. The concentration of the tracer (sulphur hexafluoride) was determined using two IR analysers for different concentration ranges, first for the range 2-30 ppm (BINOS, Leybold & Heraeus Gmbh, Germany) and second for the range 0.05-2 ppm (wavelength 10.6 µm, pathlength 21.75 m, Miran 1 A, Foxboro Inc., U.S.A.). The effect of contaminant source location on the worker's breathing zone concentration was examined using three different freestream velocities, 0.25, 0.375 and 0.5 m s^{-1}.

The use of local vertical air supply and local exhaust in reducing the transport of contaminants from the near wake region to the mannequin's breathing zone was also investigated. A local air supply unit was positioned over the mannequin (Figure 1, paper VI). Another control method studied was the local exhaust in the near wake region between the mannequin and the tracer gas release point. Different airflow rates were used.

4.5.2 Local exhaust ventilation experiments

The use of local exhaust hoods during the hand lay-up moulding of a large boat hull (length 11.5 m, total lamination area 42 m^2) was investigated in a separate lamination room (paper I). Ten exhaust ducts were located about 0.7 m over the mould surface at the centreline of the mould (figure). Based on smoke tube tests, three airflow rates (0.9, 1.4 and 2.1 $m^3 s^{-1}$) were selected for the lamination test. The efficiency of an air curtain placed between the worker and the laminate to prevent styrene exposure was also tested. Breathing zone concentrations were determined during active lamination periods. The mean consumption rate of LSE resin during the tests was 61 kg h^{-1}. The air velocities were measured with a thermoanemometer (GGA-23S, Wallac Oy, Finland). Smoke tests were again used to demonstrate airflow near the moulds.

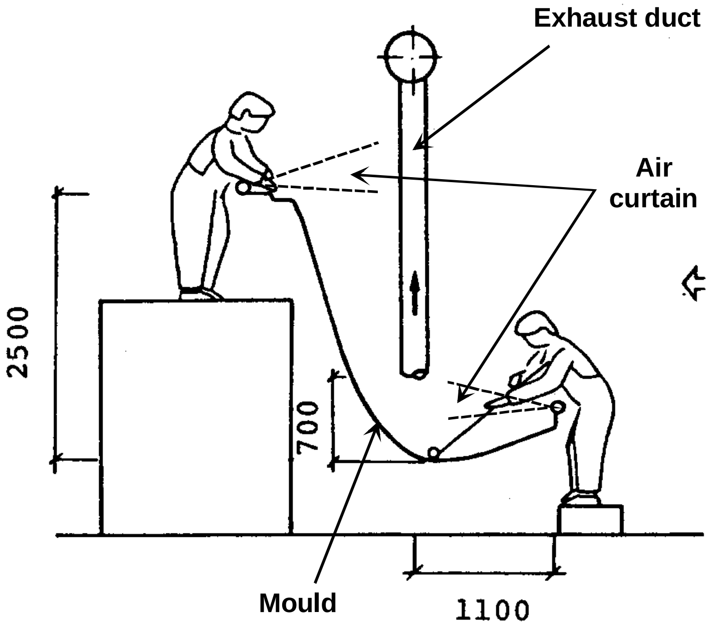


Figure 3. Location of exhaust ducts during the hand lay-up of a large boat hull.

4.5.3 Comparison of the ventilation methods during hand lay-up

The suitability of four different ventilation methods (dilution ventilation, fixed local exhaust ducts, an automatically moving local supply and a combined moving supply and exhaust) to control styrene vapours during hand lay-up moulding were compared under laboratory conditions (Section 5.3.3). Two different mould types, a plate (4.4 m × 1.2 m) and a rowboat hull (length 4.7 m), were laminated manually in a test hall (volume 100 m^2 × 6 m). The total supply and exhaust airflow rates were kept constant, 1.0 $m^3 s^{-1}$, in all four ventilation conditions. The worker's exposure to styrene was sampled into charcoal tubes on the worker's lapel during the active lamination periods. Each experiment was repeated three times, and the total number of experiments was 24. The tested ventilation configurations, airflow rates, and number of exhaust ducts used are shown in table 6.

Table 6. Tested ventilation configurations and airflow rates used in the experiments.

Configuration	**Airflow Rate $m^3 s^{-1}$**			**Number of Local Exhaust Ducts**
	Local Supply	**Local Exhaust**	**General Ventilation Supply/Exhaust**	
Dilution	0	0	1.0/1.0	0
Fixed exhausts	0	0.5	1.0/0.5	4
Moving supply	0.5	0	0.5/1.0	0
Moving exhaust & supply	0.5	0.5	0.5/0.5	2

Air was supplied to the test hall through two low-velocity air supply units (LVD-400, Halton Oy, Finland). The make-up airflow rate was 1.0 $m^3 s^{-1}$, and the temperature of the supply air was about 2°C lower than that of the exhaust air. Air was exhausted from the upper part of the hall through one exhaust opening (figure 4). The exhaust airflow rate was also 1.0 $m^3 s^{-1}$. The moulds were located under the metal cage, which was used as a frame for fixed local exhaust hoods and as a skeleton for a mobile local ventilation system.

In the fixed exhaust experiments, four exhaust openings (∅ 250 mm) were positioned over the laminate on the centre line of the mould. Local exhaust ducts were attached to the metal cage built for the mobile local ventilation system. The exhaust hoods were located as close to the laminate as possible. The distance between the laminate and the hood was 0.15 m for the plate mould and 0.5 m for the rowboat hull. The distance between the openings was 1.0 m.

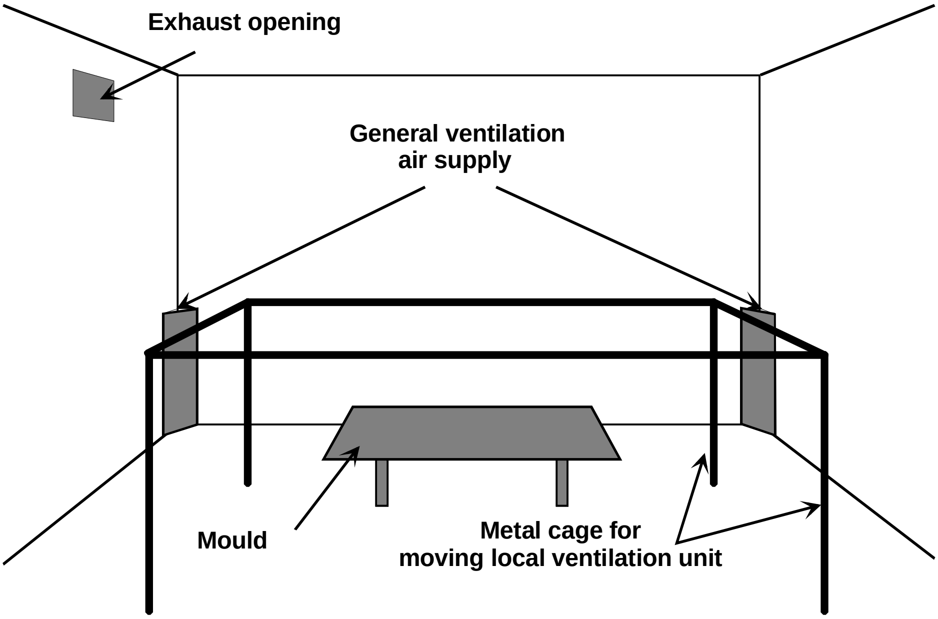


Figure 4. Dilution ventilation arrangements.

For the moving supply and moving supply and exhaust experiments, a prototype of the mobile local ventilation system was used (figure 5). It recognized the position of the person in the work area and moved the ventilation unit automatically above the worker. The ventilation unit could be equipped with a supply air diffuser or local exhaust hoods or both.

The local ventilation system consisted of a 2.5 m^2 low-velocity air supply unit faced downwards to produce a vertical clean airflow around the worker. Two unflanged circular exhaust openings were fitted to the leading edge of the air supply unit. The ventilation unit was mounted on top of a metal cage which surrounded the work area. The ventilation unit moved along tracks over the work area (figure5). The ventilated area was 6 m long and 1.5 m wide.

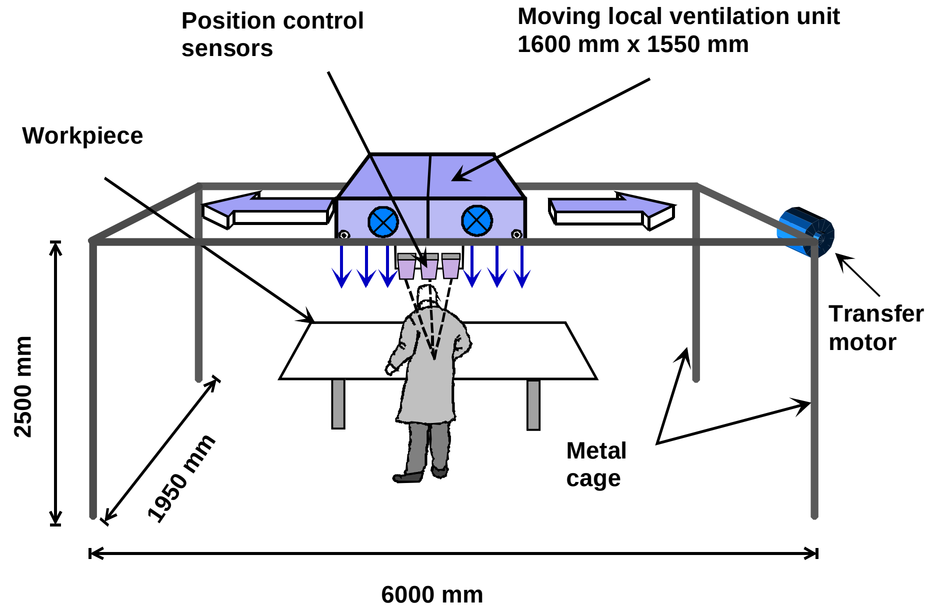


Figure 5. Principle of the automatically moving local ventilation system. The local exhaust hoods are not shown in this illustration.

The mobile local ventilation system pinpointed the position of the worker with three active IR sensors. The sensors were mounted on the rear edge of the unit, at an angle of 45^{0} pointing down towards the work area (figure 5). The IR sensors were adjusted to detect a person from a distance of about 1.5 m. The detection areas of the three sensors partially overlapped and the total detection range of the three sensors was about 1.6 m. A programmable logic unit controlled the movement of the local ventilation unit according to the signals received from the IR sensors. The variable speed drive controlled the motor moving the ventilation unit via a belt drive.

During the experiments, a moving local supply air unit was used either alone or in combination with moving local exhaust hoods. The distance between the air supply unit and the operator's breathing zone was about 1 m. The exhaust openings moved 0.15 m (plates) or 0.65 m (boat) above the laminate along the centre line of the mould. The distance between the local exhaust openings was 1.0 m. The supply and exhaust airflow rates were 0.5 m^3 s^{-1}. The temperature of the supply air was kept 2ºC lower than that of the ambient air.

Half of the plate mould (4.4 m × 0.6 m) was laminated; in other words the lamination area was 2.64 m^2. Three layers of chopped strand mat (450 g m^{-2}) were applied to the mould, and the laminate was compacted between each layer using a metal roller. LSE resin (M105 TB, Neste Chemicals, Finland) was used. The average lamination time was 31 min, and the average resin consumption rate was 14.5 kg h^{-1}. The local exhaust airflow rate per laminated area was 0.19 m^3 s^{-1} m^{-2}. During the boat hull tests, two layers of glass-fibre mat were applied to the mould. The laminate was consolidated with metal rollers. The mould was turned once during the test period so that both halves of the boat could be laminated. The average lamination time was 95 min, and the resin consumption rate was 7.5 kg h^{-1}. The total lamination area of the rowboat was about 10 m^2, about 5 m^2 of which was laminated at a time. Thus the local exhaust airflow rate per simultaneously laminated area was 0.1 m^3 s^{-1} m^{-2}.

The results were analysed with the general factorial analysis of variance (ANOVA) using the fixed effects model (procedure MANOVA, SPSS for Windows, Release 6.1.3, SPSS Inc., USA), and the comparison of the group means was performed using Duncan’s multiple range test (Montgomery 1991).

4.5.4 Improvements of the side-draught spray-up booth

The mobile local ventilation unit was also tested together with an ordinary open-front side-draught spray-up lamination booth. This study was conducted to improve the performance of the spray booth by keeping the worker under the flow of fresh air and by increasing the control velocity with movable curtains which reduced the face area of the booth. The effect of these improvements and the exhaust airflow rate of the booth (referred as factors) on the worker’s styrene exposure was investigated using the 2^3 full factorial design (Montgomery 1991) for the spraying of gel coat resin.

A spray booth (width 6.0 m, depth 2.4 m, height 2.5 m) was built in the laboratory hall. Air was exhausted through a slot on the back wall of the booth. The exhaust airflow rate was either 0.75 $m^3 s^{-1}$ or 1.50 $m^3 s^{-1}$. The mobile local ventilation unit, similar to that described in a previous section, was placed in front of the spraying booth (fugure 6). The local air supply was automatically transferred to a position above the worker, while the worker was moving laterally at the face of the spray booth. The air supply unit was constructed so that half of the unit (1.6 m × 1.6 m) was inside the booth. The fresh air supplied through the unit was 2 °C cooler than the room air, and the supply airflow rate was either 0 $m^3 s^{-1}$ or 0.5 $m^3 s^{-1}$. Mobile curtains were constructed by installing two plastic curtains on both sides of the local ventilation unit at the face of the booth. The opening between the curtains was 1.7 m (width) × 2.5 m (height). The position control system of the local ventilation unit was used to move the curtains according to the movements of the worker. The performance of the booth was examined both with and without the curtains. The factors and factor levels used in this study are shown in table. Two randomly choosen factor combinations were measured twice and the total number of of tests were 10.

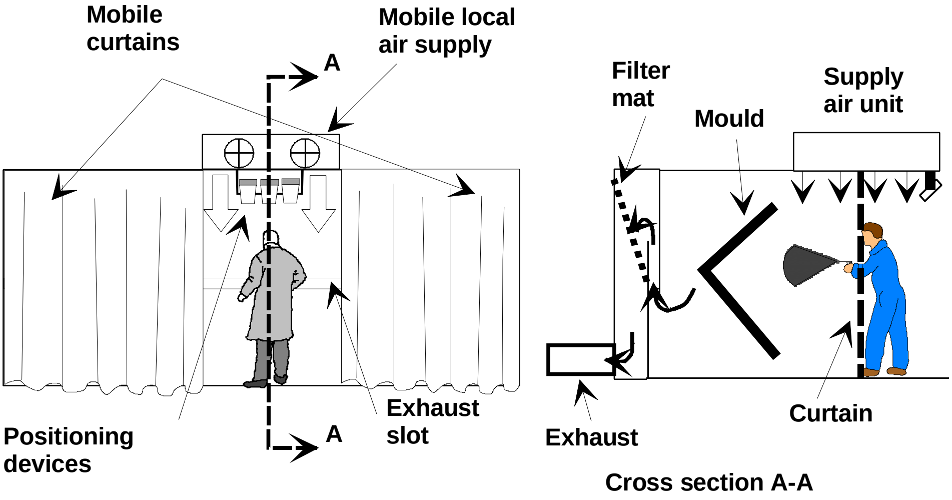


Figure 6. Schematic diagram of the improved spray booth.

Table 7. Factors and factor levels used in the spray booth experiments.

Factor	**Factor Level**	
	Low	**High**
Exhaust airflow rate	0.75 $m^3 s^{-1}$	1.50 $m^3 s^{-1}$
Supply airflow rate	0 $m^3 s^{-1}$	0.50 $m^3 s^{-1}$
Mobile curtains	No	Yes

Personal styrene samples were taken with the charcoal tube sampling method. The sampling was started just before the spraying began and was stopped immediately after the end of the spraying. The styrene concentration was also monitored outside the booth with an IR analyser. This stationary sampling point for general workroom air was located 3 m away from the face of the booth. The results were analysed with the general factorial analysis of variance using the fixed effects model (procedure MANOVA, SPSS for Windows).

During the tests, three different moulds were sprayed with gel coat resin (GE 1000 S, Neste Chemicals, Finland). The moulds were placed next to each other inside the booth. The worker started to spray the moulds from left to right, one mould at time. The worker sprayed 2.5 kg of resin onto each mould with an airless spray gun (Glas-Craft LPA-III-AAC, Indianapolis, IN, USA). The spraying parameters were kept constant during the tests.

5. Results

5.1 Emission of styrene

5.1.1 Emission of styrene from standard and low emission resins during hand lay-up

The styrene evaporation varied between 2.1% and 6.9% of the styrene content of the resin, depending on the resin type and the laminate thickness (Paper III). The total evaporation was 30-60% lower with LSE resin than with standard resin. The emission rate varied strongly with time, being highest during rolling (Figure 2, Paper III). In this phase, the difference in the emission rate between LSE and standard resin was only 4-14%. As the lamination was finished, the emission rate decreased rapidly for the LSE resin but continued intensively for the standard resin. In the curing phase, the styrene emission again increased notably for the LSE resin, as the heat generation inside the laminate began due to exothermic polymerization. This phenomenon was significant, especially with thick (12 layers) laminates. With standard resin, the increase in the emission rate was almost imperceptible during the curing phase.

The study indicated (Figure 3, Paper III) that both resin types released about the same amount of styrene during rolling. However, compared with the standard resin, the LSE resin had a lower evaporation rate during the period between the rolling and curing. This fact explains the smaller total mass of styrene evaporated from the LSE resin.

5.1.2 Effect of rolling area on styrene emission

The emission rate of styrene during the rolling of LSE resin depended significantly on the amount of simultaneously rolled area (oneway ANOVA, $p<0.001$). The emission rate from the mould was highest when the whole surface area was rolled (figure 7). The lowest emission rate was detected when only a quarter of the mould (0.75 m^2) was rolled out at a time. All the mean values were significantly different (Duncan’s multiple range test, $p<0.05$).

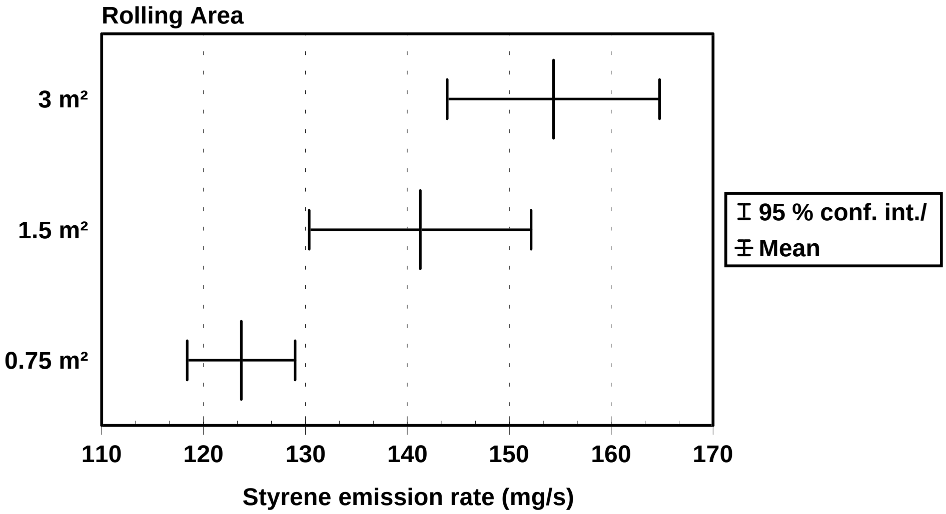


Figure 7. Effect of rolled surface area on the styrene emission rate.

5.2 General ventilation

5.2.1 Evaluation of the ventilation methods used in Finnish reinforced plastics plants

The workers’ styrene exposures were generally high in the 17 plants studied (Paper II). About 70% of time-weighted average (8h) styrene concentrations (n=105) measured in the laminator’s breathing zone exceeded the present Finnish occupational exposure limit of 20 ppm. One-third of the breathing zone concentrations were even higher than 50 ppm. About 45% of the area samples exceeded 20 ppm. The styrene concentrations were notably higher in the breathing zone than in the background area, 1-7 m away from the worker (figure 8). The styrene concentration had to be less than 10 ppm in the general workroom air prior to the 20 ppm exposure level was achieved.

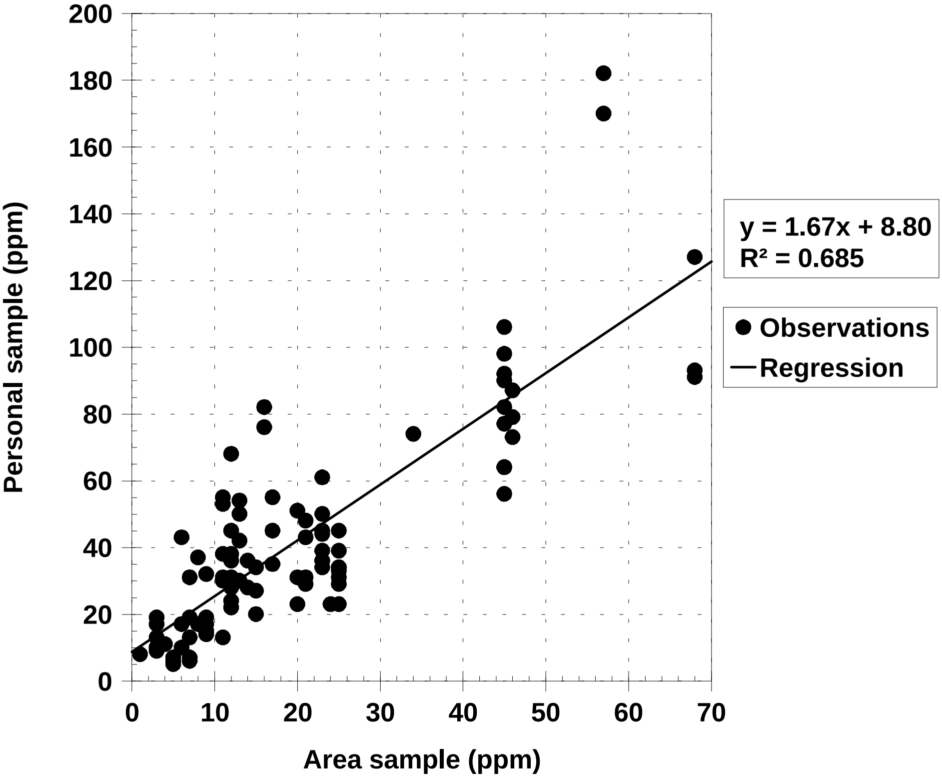


Figure 8. Regression between personal concentrations and background area concentrations.

The air change efficiency (ε_a) ranged from 0.47 to 0.63 for the lamination rooms; therefore the supply air was rather well mixed with the room air (ε_a about 0.5) and, in plants utilizing recirculating air jets, a tendency towards displacement flow (ε_a =0.57-0.63) was observed. The dilution air volume per resin mass varied between 200 and 1200 $m^3 kg^{-1}$. In Figure 6 of Paper II, the air-to-resin ratio was compared with the breathing zone concentrations measured in the plants. The mean values were below 20 ppm for four plants. Two of them used the closed moulding method. In the third plant, using the filament winding method, the dispersion of styrene was controlled with enclosures, local exhaust ventilation and isolation of different work operations. High airflow rates in relation to resin consumption was used in the fourth plant, using hand lay-up moulding. In this plant, unidirectional airflow was achieved in the lamination area ($\varepsilon_a = 0.57$), the work practices were synchronized with the airflow pattern, and a large airflow rate, 850 $m^3 kg^{-1}$, was used.

Figure 6 of Paper II indicates that high air volumes per resin mass were needed to control the styrene concentration with dilution ventilation. Especially at higher airflow rates, a notable increase in the dilution flow rate caused only a minor decrease in the concentration of styrene. The dilution airflow rates were too low in many plants. In 8 plants out of 15 using hand lay-up and spray-up methods, the styrene concentrations, calculated from the measured styrene emission and the airflow, even exceeded 20 ppm. This finding indicated that it was not possible, even in theory, to attain compliance with the occupational exposure limit.

5.2.2 Use of zonal unidirectional airflow

Exposure measurements performed in the polyester plastics plants using recirculation air jets indicated that the use of horizontal unidirectional airflow with a velocity of about 0.3 $m s^{-1}$ over the mould can reduce styrene concentrations to below 50 ppm (Table I - Table III, Paper I). The level of 20 ppm is, however, difficult to achieve. This kind of unidirectional airflow pattern can be created by the use of recirculation air jets as in plant 1 (Figure 3, Paper I) and in a small boat lamination area in plant 2 or by the use of direct make-up airflow in plant 2 in the lamination area as in the large boat lamination area.

The position of the mould in the airflow proved to be important (Table I, Paper I). It is a common practice that two workers laminate together on large moulds (>7 m). It was natural that the worker standing downwind was more exposed than the worker upwind when the mould was placed parallel to the airflow. The perpendicular position of the mould was more favourable because both workers stayed in front of the emission source in relation to the make-up airflow.

A horizontal unidirectional airflow was achieved in two tunnel-like lamination rooms (Paper IV). The air change efficiencies were from 0.78 to 0.85 in both lamination rooms (Table 1, Paper IV). The local air change indices measured in the middle of lamination room II varied from 1.23 to

1.40, indicating a displacement type airflow pattern (Table 2, Paper IV). The contaminant removal effectiveness within the lamination room was dependent on the injection site (Table 3, Paper IV). The contaminant removal effectiveness was below 1.00 (i.e., the value of complete mixing) near the stagnation eddies due to the vertical obstacles of the mould. The effectiveness was greater than 1.00 for the horizontal mould surfaces, and therefore fast evacuation of the contaminated air was indicated.

5.3 Local ventilation

5.3.1 Formation and control of wake near the worker

The experiments with an isothermal mannequin in horizontal unidirectional airflow showed that a three-dimensional wake region formed downstream of the mannequin, and significant contaminant transport towards the mannequin's breathing zone occurred in this recirculating wake area (Paper V). The transport of the contaminants depended on the height and the distance of the contaminant source from the body and on the distance from the vertical mid-section (Figure 3, Paper V). The exposure decreased rapidly when the distance between the worker and the contaminant source increased. Downwash was also observed as the air flowed over the mannequin's head. Contaminant transport toward the breathing zone occurred above the mannequin's hip level (height 0.8 m), but contaminants released below this level did not significantly enter the breathing zone (Figures 3 and 5, Paper V). The length of this contaminant transport region was about 0.5-0.6 m, and it was not affected much by the freestream velocity (Figure 5, Paper V).

The exposure caused by the wake could be reduced by the use of local exhaust positioned in the wake region or by a local air supply positioned over the mannequin, the later method being more efficient (Paper VI). With supply airflow rates greater than 0.075 $m^3 s^{-1}$, the measured tracer gas concentrations were lower than 0.2 ppm in the breathing zone, less than 2% of the reference concentration, from all the contaminant release points. The use of local air supply notably reduced the size of the region where significant contaminant transport occurred (Figure 4, Paper VI).

The benefits of the local air supply were clearly seen also when the mean exposure caused by the release of the contaminant from the area representing the work area was studied (Figure 6, Paper VI). An air supply of 0.075 $m^3 s^{-1}$ reduced the exposure to one tenth of that without any air supply.

The experiments done with the exhaust duct positioned near the worker showed that the wake region was unstable and sensitive to disturbances. The addition of the exhaust duct to the wake region reduced the average relative exposure by one half even though there was no exhaust airflow (Figure 7, Paper VI). An exhaust airflow of 0.15 $m^3 s^{-1}$ further reduced the exposure by 50%. However, a local air supply was more effective in controlling the exposure caused by the contaminant source in the wake region near the worker (Figures 6 and 7, Paper VI).

5.3.2 Local exhaust ventilation for the hand lay-up of large parts

The use of a large number of exhaust ducts (10 ducts) reduced the styrene concentrations notably in the worker's breathing zone during the hand lay-up moulding of a large boat hull (length 11 m) (Table IV, Paper I). It was possible to control the styrene concentration to near 20 ppm using a relatively high exhaust airflow rate, 2.1 $m^3 s^{-1}$ (about 0.05 $m^3 s^{-1} m^{-2}$). The air velocities generated by local exhaust ducts varied from 0.15 to 0.25 $m s^{-1}$ in front of the mould. The advantage of an air curtain between the worker and the laminate proved to be marginal. Although the air curtain increased the average air velocities over the mould, it generated styrene dispersing eddies over the mould at the same time.

5.3.3 Comparison of different ventilation systems

As expected, dilution ventilation (air-to-resin ratio 280 $m^3 kg^{-1}$ for plates and 480 $m^3 kg^{-1}$ for boats) was not capable of keeping the styrene concentration level below 20 ppm in the laminator's breathing zone during the hand lay-up tests in the laboratory. The use of local ventilation methods reduced the exposure significantly (table 8). With fixed local exhaust ducts the concentration of styrene was near the 20 ppm level in the breathing zone (figure 9), and was 40% to 56% lower than when dilution ventilation was used. The airflow rate of the exhaust ducts, when compared with that of the lamination area, was 0.19 $m^3 s^{-1} m^{-2}$ during plate lamination and 0.1 $m^3 s^{-1} m^{-2}$ during boat hull lamination. The difference between the dilution ventilation and fixed exhaust hoods was statistically significant (Duncan’s multiple range test, p=0.05).

Table 8. Analysis of variance for comparing the ventilation systems. The dependent variable was the styrene concentration in the worker’s breathing zone.

Source of Variation	**Sum of Squares**	**D.F.**	**Mean Square**	**F_0**	**F Probability**
Mould type	133.81	1	133.81	5.70	0.030
Ventilation method	2296.57	3	765.52	32.58	<0.001
Mould and Ventilation	362.53	3	120.84	5.14	0.011
Error	375.91	16	23.49		
Total	3168.83	23	137.78		

The interaction between the mould type and ventilation method can be seen in table 8 for moving supply ventilation and moving supply and exhaust ventilation. The use of a mobile local ventilation unit with a local air supply decreased the exposure to clearly below the 20 ppm level during plate lamination (figure 9). With this type of ventilation the worker’s exposure was about 80% lower than with the dilution ventilation; it was also significantly lower than with the use of fixed local exhaust hoods (Duncan’s test, p=0.05). However, there was no difference in exposure when the exhaust ducts were combined with a mobile local air supply unit. During rowboat lamination the mobile local ventilation unit was not as effective as during plate lamination. There was no difference in the styrene concentration when the mobile ventilation unit was used with the air supply alone or together with exhaust openings when compared with that of fixed local exhaust hoods (Duncan's test, p=0.05). Styrene exposure during rowboat lamination was near or slightly over 20 ppm when local ventilation methods were used. The reduction of the worker's exposure with different local ventilation methods during rowboat lamination varied between 30% and 56% when these methods were compared with dilution ventilation.

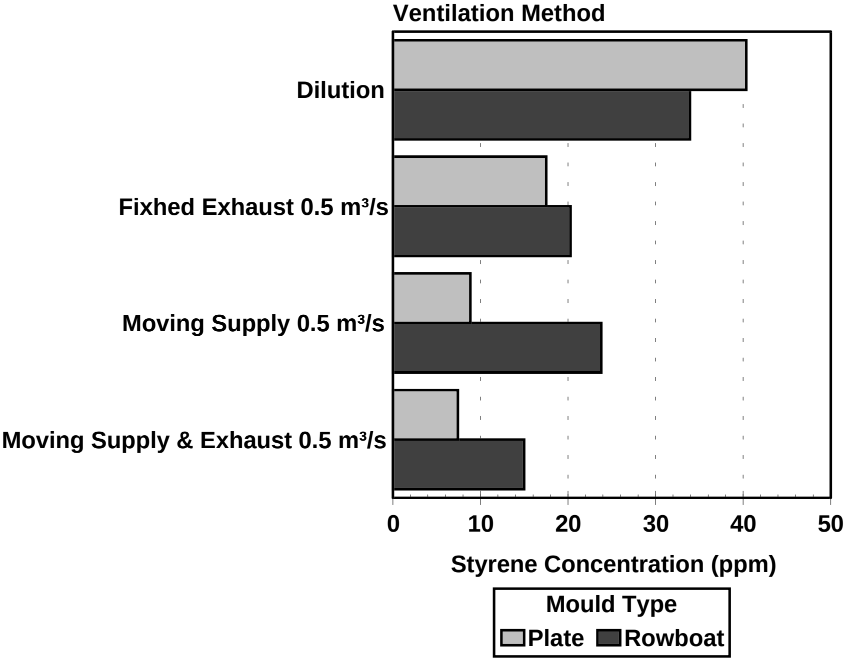


Figure 9. Comparison of the average styrene concentrations with different ventilation methods during hand lay-up moulding of rowboats and plates.

5.3.4 Improved spray-up booth

As expected, the worker's exposure was high, varying between 100 and 280 ppm in the gel coat spraying experiments with the conventional open-faced spraying booth. The theoretical face velocities obtained with the exhaust alone varied between 0.05 and 0.1 m s^{-1} and they were too low to control the styrene emission during the spraying.

The effect of the exhaust airflow rate, the use of local air supply and the use of mobile curtains on the worker's exposure were analysed with an analysis of variance (table 9).

Table 9. Factorial analysis of variance for the spray booth improvement tests. The dependent variable was the styrene concentration in the worker's breathing zone.

Source of Variation	**Sum of Squares**	**D.F.**	**Mean Square**	**F_0**	**F probability**
Curtains	38 598.32	1	38 598.32	16.39	0.007
Exhaust flow rate	16 489.83	1	16 489.83	7.00	0.038
Supply air	23 435.54	1	23 435.54	9.95	0.020
Curtain and Exhaust	927.77	1	927.77	0.39	0.553
Curtain and Supply	425.69	1	425.69	0.18	0.686
Exhaust and Supply	12 636.33	1	12 636.33	5.36	0.060
Error	14 134.01	6	2 355.67		
Total	104 848.90	12	8 737.41		

When the exhaust airflow rate was increased, mobile curtains were used to reduce the open face area, and a mobile local air supply unit was placed over the worker, the worker's exposure decreased notably (figure 10). The increase in the exhaust flow rate from 0.75 to 1.5 $m^3 s^{-1}$ reduced exposure by an average of 65%. The use of mobile curtains also markedly decreased the styrene concentration in the worker's breathing zone (figure 10). Styrene exposure was about 60% lower when the curtains were used. The open face area of the booth was reduced from 15 to 4.25 m^2, giving the theoretical control velocities of 0.18 m s^{-1} for 0.75 $m^3 s^{-1}$ and 0.35 m s^{-1} for 1.5 $m^3 s^{-1}$. In addition, the use of a mobile air supply unit also affected the exposure, but the effect was divaricate (figure 10). With a low exhaust airflow rate, the local air supply decreased the exposure about 65%. Surprisingly, the air supply did not affect the exposure when the exhaust airflow rate was high. This result did not concur with the original hypothesis.

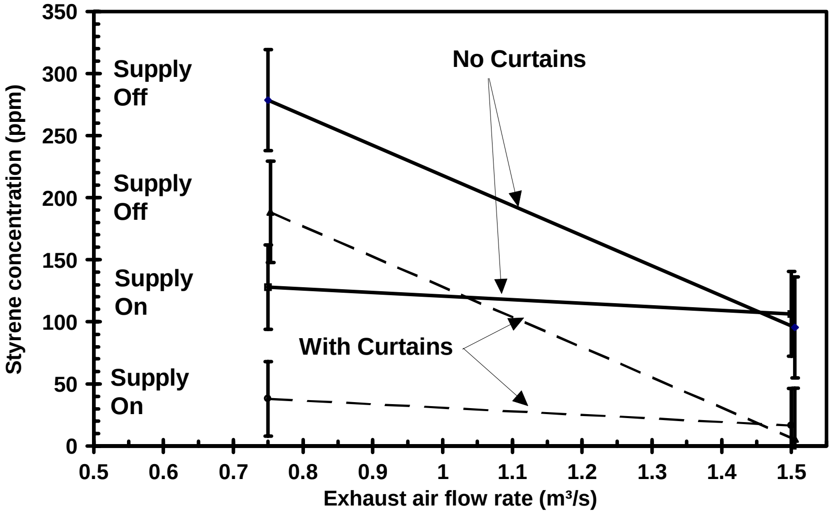


Figure 10. Effect of the exhaust flow rate, air supply and curtains on the styrene concentration in the worker's breathing zone during the spraying of gel coat resin. The vertical bars show the 95% confidence intervals of the predicted means.

The concentration of styrene remained below the 20 ppm level when a high exhaust airflow rate was used together with mobile curtains. This combination gave a calculated control velocity of 0.35 m s^{-1} when based on a free opening and exhaust airflow rate. The use of a local air supply decreased the exposure only when the low exhaust airflows was low.

The mobile curtains, the air supply unit, and the exhaust airflow rate affected the general air styrene concentration in the room outside the booth. Only the main effects were statistically significant (ANOVA, $p<0.05$). The doubling of the exhaust airflow rate had the greatest effect. The general styrene concentration was about 90% lower when a high exhaust airflow rate was used. The mobile curtains reduced the styrene concentration by 40%, whereas the use of a mobile air supply increased the styrene concentration in the general workroom air. When the air supply was used, the styrene concentration was 60% higher in the general workroom air than without it.

6. Discussion

6.1 Emission of styrene

In this study, the use of LSE resin decreased the evaporation of styrene by 30-60% (Paper III). These values are somewhat lower than the reductions reported by Andersson et al. (1994), 55-70%. The use of LSE resin has been reported to reduce styrene concentrations by 27% to 50% in laminators' personal air samples (Kalliokoski et al. 1984, Pfäffli et al. 1992, Andersson 1995).

The evaporation of styrene was greatest during the rolling process (Paper III) as has also been detected by Pritchard & Swapillai (1978) and Andersson et al. (1994). In the present study, the styrene emission rate was only somewhat lower with LSE resin than with standard resin during rolling (Paper III). The purpose of vapour suppressants is to build a diffusion barrier on the surface of the laminate. However, the additives are mixed back into the resin during the rolling, and no barrier can be formed. Only after rolling can the barrier be established. This phenomenon is also indicated by the low evaporation of styrene from LSE resin during the period between the rolling and curing (Paper III). This phase accounts for most of the difference in total styrene evaporation between LSE and standard resins (Paper III).

In practice, the entire mould area is not usually rolled at the same time. The emission rate of styrene was found to depend on the rolled surface area (Section 5.1.2). The greater was rolled area, the higher was emission rate. The vapour suppressant additives covered part of the surface when large areas were rolled. Therefore, the use of LSE resin probably decreases styrene emission, and thus the styrene exposure during the rolling phase of large products, but the reduction may be minimal when small products are laminated.

6.2 Control of air currents near the worker and the source

6.2.1 Dilution and zonal unidirectional flow ventilation

In practice, all of the 17 plants studied used dilution ventilation to control styrene exposure (Paper II). Many of the ventilation systems were poorly designed. The airflow rates were too low, even theoretically, to provide satisfactory exposure control. Therefore the exposure to styrene was high and the Finnish occupational exposure limit was often exceeded (Paper II). This finding agrees with the findings of Pfäffli et al. (1992), who found that 77% of lamination workers (n=248) were exposed to levels higher than 20 ppm.

One important reason for high styrene exposure in the present study was work in the immediate vicinity of the emission source (Paper II). Impractically high airflow rates, over 850 m^3 per kilogramme of resin, are needed to keep styrene exposure below 20 ppm with dilution ventilation during hand lay-up moulding (Paper II). In addition, the styrene concentration in the general workroom air should be below 10 ppm level. In plants using closed mould methods, styrene emission is low and the workers do not need to stay continuously in the immediate vicinity of the emission sources; therefore dilution ventilation may be an adequate control method under these circumstances (Kalliokoski et al. 1994). In hand lay-up moulding with a resin consumption rate of 20 kg h^{-1}, controlling the styrene concentration with dilution ventilation is difficult, and, with a very high consumption rate, more than 100 kg h^{-1}, it is no longer a real alternative. Crandall (1981) suggested that, in some instances, dilution ventilation is a satisfactory method for exposure control. This assumption may be true, especially, if the target level is as high as 50 ppm. But, generally, dilution ventilation needs to be merged with other more-sophisticated control measures such as styrene-suppressed resins, zonal ventilation, local ventilation, and work practice guidelines.

In some cases, the performance of general dilution ventilation can be improved by creating zonal unidirectional airflows. High-velocity recirculation air jets are one possible means of creating a unidirectional airflow zone within the lamination room. Air change efficiency in such plants has been found to range from 0.57 to 0.64, indicating a slight tendency towards displacement flow (Paper II, Niemelä et al. 1992). However, this kind of ventilation system was able to control styrene exposure only below 50 ppm during the hand lay-up moulding of both small and large boats with normal resin consumption rates (Paper I and II). Similar results have been reported for systems producing a horizontal airflow over the hull mould (length 14 m) according to the push-pull ventilation principle (Todd 1985). If the exhaust systems are not properly designed, systems of this kind tend to spread styrene vapours rather than remove them. However, if the airflow rate is high in comparison with the resin consumption (over 850 $m^3 kg^{-1}$) and the designer of the system has ensured that the air flows in such a way that the employees receive uncontaminated air, styrene exposure levels below 20 ppm may be achieved (Paper II).

A more effective vertical displacement flow pattern can be achieved by introducing an air supply through a large perforated plenum and exhausting it through a large air outlet in the opposite wall. The airflow pattern was more unidirectional and the air change efficiencies measured in these plants were higher than in plants using booster jets to transport make-up air (Paper IV). The contaminant removal effectiveness was mostly greater than 1.00 in these plants and therefore indicated good transportation of contaminants towards the exhaust. The capability of the system to control styrene vapour has also been confirmed by exposure measurements. Andersson et al. (1993a) showed that a worker's exposure remains well below 20 ppm during the lamination of a 5 m long boat part when only a moderate airflow rate is used in comparison with the resin consumption (620 $m^3 kg^{-1}$). Better air quality is attained using less make up air with this kind of lamination tunnel than with the conventional dilution ventilation.

Unidirectional airflow alone may not suffice to produce good air quality in the worker's breathing zone. Proper work practices in synchrony with the airflow pattern is crucial (Paper II). The effect of unidirectional airflow on a worker's exposure depends on the worker's location in relation to the styrene source and the fresh air stream. A low level of styrene exposure can be attained only when a worker moves toward the fresh airflow and the evaporating surface stays downstream (Paper I). These findings also agree with those of previous studies concerning the effect of a worker's position relative to the airflow on exposure (Flynn & Shelton 1990, George et al. 1990, Andersson et al. 1993a). These facts indicate that the combination of good ventilation and a correct approach to work practices gives the best results in reducing exposure to styrene.

6.2.2 Air currents near the worker in unidirectional airflow

Many of the ventilation controls used in GRP plants, such as spray or lamination booths and horizontal flow lamination tunnels, form a unidirectional airflow field around a person. The use of these controls may, however, have problems. Several authors have shown that the person standing in an airflow can become significantly exposed due to a recirculating airflow being created downstream of the body (Ljungqvist 1979, 1987, George et al. 1990, Kim & Flynn 1991a, 1991b, Flynn & Ljungqvist 1995). The present study showed that the flow field downstream of a mannequin is very complex and, as a result, the transport of the contaminants depends greatly on the location of the source. At the velocity range studied, the length of the recirculation flow depended very little on the freestream velocity, and it appeared to be about 1.5 times the mannequin's width. It was also observed that significant contaminant transport toward the breathing zone occurs only above the hip level, due to two eddies formed in the wake region. The upper eddy recirculated upwards near the worker, while the lower one recirculated in the opposite direction (Paper V). Thus the upper recirculation zone was the most important region with respect to a worker's exposure. These findings are in general agreement with those of previous studies (George et al. 1990, Kim & Flynn 1991a). However, all these studies were done using an isothermal mannequin. Recent studies have demonstrated that thermal effects may be important to near-body airflow field (Johnson et al. 1996).

The wake region is sensitive to disturbances. Kim and Flynn (1992) discovered that a source momentum or a flat plate downstream of the worker clearly reduces the breathing zone concentration. In the present study, the exhaust duct located in the wake region near the worker reduced exposure without any exhaust air (Paper VI). It was also shown that the increase in the freestream velocity was not an efficient control measure (Paper V). These findings indicate that the flow in the wake region can easily be affected and effective local control measures can be produced.

When the air was exhausted from the wake region at a rate of 0.150 $m^3 s^{-1}$, the exposure was reduced about 50% (Paper VI). However, when an air supply over the mannequin was used with the flow rate of 0.1 $m^3 s^{-1}$, the mean breathing zone concentration was reduced over 99% (Paper VI). In this situation, an supply air velocity of 0.28 $m s^{-1}$ was high enough to control the exposure; this finding agrees with the results of Gressel and Fischbach (1989). The present study suggests that the use of local supply air can efficiently control the contaminant transporting eddies generated in front of a person.

6.2.3 Fixed local ventilation methods for hand lay-up

Local exhaust ventilation was tested in one GRP plant (Paper II) and in the laboratory (Section 5.3.3). The airflow velocity over the mould should be high enough to overcome disturbing air currents. This study showed that the control velocity should be at least 0.2 $m s^{-1}$ in hand lay-up moulding (Paper I). This value is at the lower end of the control velocities normally recommended for evaporating liquids (ACGIH 1992). On the other hand, Fletcher and Johnson (1986) have also found the control velocity of 0.2 $m s^{-1}$ to be high enough to capture contaminants from a low velocity source. It was also observed in the present study that sufficient control of styrene can be achieved if the exhaust airflow rate per laminated area is on the level of 0.1 to 0.2 $m^3 s^{-1} m^{-2}$, depending on the mould shape (Paper I, Section 5.3.3). Concave moulds, such as boat hulls, are more favourable for local exhaust ventilation than flat or convex moulds are. The exhaust airflow rates used in this study are much lower than the exhaust values recommended for similar operations in large open areas, such as 0.62-0.85 $m^3 s^{-1} m^{-2}$ for dip tanks (ACGIH 1992). Maintaining styrene exposure at a level below 20 ppm has been previously achieved when the local exhaust airflow rate varied between 0.1 and 1.0 $m^3 s^{-1} m^{-2}$ (Isaksson 1976, Ivalo et al. 1985).

The drawback of local exhaust is its very short effective control range. Ivalo et al. (1985) estimated that about one exhaust hood per square metre of laminate is needed to control styrene exposure during hand lay-up moulding. Therefore many exhaust hoods are needed to control large moulds and they may disturb the work. One possibility for enlarging the control range of the exhaust hood would be to combine them with an air curtain over the laminate. However, this study showed that the effect of an air curtain is marginal (Paper I). The air curtains generated eddies over the mould and increased the exposure on the opposite side of the mould.

Another difficulty with exhaust ventilation is the need to design the location of the exhaust hoods individually for every mould type. The difficulties with local exhaust ventilation were seen in the few plants in which flexible hoses were installed. The exhaust ducts were not located properly, and they functioned almost as exhausts for general ventilation (Paper II).

6.2.4 Mobile local ventilation system

A mobile local ventilation system, which detects the position of the worker and automatically moves the ventilation unit above the laminator, was effective in controlling the styrene exposure from medium-size laminates. During the hand lay-up moulding of a 4.4 m long plate and a 4.7 m long rowboat hull, it was possible to achieve breathing zone styrene concentrations below or at least near the 20 ppm level (Section 5.3.3). The system turned out to be at least as effective as the fixed local exhaust hoods. The mobile system can be used to replace fixed local exhaust systems with many hoods.

The performance of the system was dependent on the shape of the mould. The mobile system was efficient for plates. In this case, the local air supply reduced the laminator's styrene exposure more than the fixed exhaust ducts did. Adding moving local exhaust hoods did not affect the exposure, but it may have reduced the amount of styrene vapours escaping to the general workroom air and thus reduced the exposure of non-laminators. In both cases, the worker's exposure was below 10 ppm. However, the local air supply was not always effective. In the case of boat lamination, the mobile ventilation unit did not affect the exposure when compared with the fixed exhaust hoods. The styrene concentration in the worker's breathing zone remained between 15 and 25 ppm. The difference in the workers' styrene exposure during plate and boat lamination was unexpected. It may have been due to the different work postures required in these jobs. During the lamination of plates, the worker was able to stand straight. On the other hand, when he worked with the rowboat hull he had to bend forward. This bent posture, with the airflow from the supply unit located over the worker, may have produced eddies in front of the worker and thus resulted in the transport of styrene vapours toward the breathing zone. Other recent studies have also given some evidence that a bent work posture may increase the exposure level when local air supply is used (Enbom 1989, Säämänen 1997).

The average air supply velocity was only about 0.2 m s^{-1} at the plenum, and this level is lower than velocities commonly used (Cecala et al. 1988, Volkwein et al. 1988, Gressel & Fischbach 1989). The use of a low velocity was possible because the temperature of the local air supply was kept slightly cooler than that of the room air. The buoyancy force of the cool air supply assists the inertia forces and increases the momentum of the air supply, the result being a greater amount of fresh air in the worker's breathing zone than in the case of isothermal air. In most previous studies, isothermal air has been introduced to the air supply device. Therefore high air velocities should be used to ensure good downward air movement.

The prototype of the mobile local ventilation system focused the exposure control on the area where it was most needed. The local exhaust hoods of the system are always located as near the emission source as possible because the rolling of the laminate generates the highest emission, while the emission from the rest of the mould is much lower. In addition, instead of creating an entire ceiling of clean air diffusers, the mobile local air supply unit is moderate in size.

6.2.5 Spray booth with mobile curtains and a local air supply unit

Several methods were used to improve the performance of the conventional open-front side-draught spray booth (Section 5.3.4). Both an increase in the exhaust airflow rate and the use of mobile curtains decreased the worker's exposure during gel coat spraying. Each of these measures increased the control velocity at the face of the booth. The lowest exposure, below 20 ppm, was detected when the control velocity was 0.35 $m\ s^{-1}$. This velocity is at the lower end of the values (0.3-1.5 $m\ s^{-1}$) recommended for airless spray gun operations in large spray booths (Fletcher 1989, ACGIH 1992). Isaksson (1976) has shown that the styrene concentration can be kept below 20 ppm in the breathing zone of the spray gun operator if the control velocity of the booth is between 0.3 and 0.5 $m\ s^{-1}$ during the spraying.

Isaksson (1976) also used curtains at the face of the spray booth to increase the enclosure degree and to reduce the exhaust airflow rate required. The weakness of his system was the need to move the curtains manually. In the present system, the curtains were moved automatically. The same system that moved the local ventilation unit also moved the curtains. This procedure reduced the exhaust airflow rate needed to keep the styrene concentration at a moderate level and relieved the worker from the responsibility of having to move the curtains manually. Because of the relatively low exhaust airflow rates (0.75-1.5 $m^3\ s^{-1}$) used in this study, it was not possible to reach a styrene level below 20 ppm in the worker's breathing zone without curtains. Comparable results have been obtained in Sweden, when an automatically moving canvas-covered frame was used at the face of the spray booth (Andersson & Rosén 1994).

The effect of the mobile local air supply was slightly unexpected. Measurements with the mannequin in a unidirectional airflow (Papers V, VI), suggested that a mobile local air supply over the worker would decrease the exposure markedly. However, the results showed that the local air supply was effective only when the exhaust airflow rate was low (0.75 $m^3\ s^{-1}$). Air currents that differed from the ones expected between the mould and the worker may have led to this discrepancy. The converging flow into the spray, the momentum of the spray gun and the moulds inside the booth may have altered the formation of the reverse flow eddies downstream of the worker (Kim & Flynn 1992, Dunnet 1994, Flynn et al. 1996) and therefore altered the expected styrene reduction. Having a local air supply at the face of the booth may have also disturbed the flow into the booth to some degree, as indicated by the elevated styrene concentrations in the general workroom air. After all, the results indicated that, with the mobile local air supply, the worker's exposure to styrene was lower or equal to that without the air supply.

7. Conclusions

Dilution ventilation systems are commonly used in Finnish glass-fibre reinforced polyester plastic plants to control styrene vapours. In many cases, the dilution ventilation is incorrectly dimensioned and workers are exposed to high styrene concentrations. If open moulding methods are used, impractically high dilution airflow rates, over 850 $m^3\ kg^{-1}$, are needed. Therefore, it is unlikely that concentrations below the exposure limit of 20 ppm can be achieved with dilution ventilation at the rates of resin consumption common today.

The use of vapour-suppressed resin reduces the total styrene emission, but the reduction of exposure achieved with low styrene emission resins is lower than expected according to the total emission tests. Vapour-suppressant additives reduce emission markedly only when the laminate surface is not rolled and, therefore, vapour-suppressed resins are beneficial when large laminates are manufactured. The use of low styrene emission resins is one of the key components of styrene control in reinforced plastics plants.

Unidirectional airflow in the lamination zone created with the high-velocity air jets did not keep the styrene exposure level below 20 ppm. With this system, low exposure levels require that very high make-up airflow rates, over 850 $m^3\ kg^{-1}$, be used. A more effective airflow pattern can be achieved in a horizontal flow lamination tunnel. With this system, it is possible to keep the styrene exposure level clearly below the current occupational exposure limit of 20 ppm with a moderate (about 650 $m^3\ kg^{-1}$) make-up airflow rate. In unidirectional airflow systems, work practices should be synchronized with the airflow pattern.

A worker standing in a unidirectional airflow facing downstream can receive significant exposure due to contaminant transporting eddies formed downstream of the body. The flow field within the wake is very complex, and the transport of contaminants depends on the location of the source. The recirculation airflow area is long enough to transport contaminants from hand-held sources to the worker’s breathing zone. It is possible to control the exposure with a local air supply unit located over the worker. Only a relatively low supply airflow rate is needed to control the worker's exposure in such cases.

The commonest method of controlling the emission of airborne contaminants is the use of local exhaust ventilation. The control velocity at the edge of the mould should be at least 0.2 m s^{-1} during hand lay-up moulding. It was estimated that the adequate styrene control can be achieved when the airflow rate per laminated area is between 0.1 and 0.2 m^3 s^{-1} m^{-2}. Several exhaust hoods are needed for large moulds. Using an air curtain over the mould to reduce the exhaust airflow rate is not beneficial.

An automatically moving local ventilation unit can be used to control larger moulds with moderate airflows during hand lay-up moulding. The styrene concentration can be near or below 20 ppm when a mobile air supply is used in combination with mobile exhaust hoods. This system can be used to replace fixed local exhaust systems with many hoods, at least when medium-size objects are laminated.

A new type of spray booth, equipped with automatically moving curtains and a local air supply, can reduce the worker's exposure to styrene markedly during the spraying of polyester resin. Styrene concentrations below 20 ppm can be obtained when the open face area of the booth is reduced with curtains and the control velocity is at least 0.3 m s^{-1}. The use of a local air supply reduces the exposure when the control velocity is low.

Control of styrene vapours in reinforced polyester plants is difficult because the work is mainly done manually with open moulds. Successful exposure prevention can be obtained if the emission of styrene is kept as low as possible. In addition, the airflow field near the worker has to be carefully designed. Zonal ventilation methods, such as horizontal airflow tunnels, and novel local ventilation systems can be used to control laminators' styrene exposure below the occupational exposure limit.

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