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**Original Article**

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## Work Environments and Exposure to Hazardous Substances in Korean Tire Manufacturing

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**Objectives:** The purpose of this study is to evaluate the tire manufacturing work environments extensively and to identify workers' exposure to hazardous substances in various work processes.

Methods: Personal air sampling was conducted to measure polycyclic aromatic hydrocarbons, carbon disulfide, 1,3-butadiene, styrene, methyl isobutyl ketone, methylcyclohexane, formaldehyde, sulfur dioxide, and rubber fume in tire manufacturing plants using the National Institute for Occupational Safety Health Manual of Analytical Methods. Noise, carbon monoxide, and heat stress exposure were evaluated using direct reading instruments. Past concentrations of rubber fume were assessed using regression analysis of total particulate data from 2003 to 2007, after identifying the correlation between the concentration of total particulate and rubber fume.

Results: Workers were exposed to rubber fume that exceeded 0.6 mg/m<sup>3</sup>, the maximum exposure limit of the UK, in curing and production management processes. Forty-seven percent of workers were exposed to noise levels exceeding 85 dBA. Workers in the production management process were exposed to 28.1°C (wet bulb globe temperature value, WBGT value) even when the outdoor atmosphere was 2.7°C (WBGT value). Exposures to other substances were below the limit of detection or under a tenth of the threshold limit values given by the American Conference of Governmental Industrial Hygienists.

**Conclusion:** To better classify exposure groups and to improve work environments, examining closely at rubber fume components and temperature as risk indicators in tire manufacturing is recommended.

**Key Words:** Rubber, Occupational exposure, Aerosols, Carbon disulfide

## **Introduction**

Seven cases of sudden cardiac death and five cases of cancer occurred among the employees of a particular Korean tire manufacturer between May 2006 and September 2007, leading

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to concerns about the work environments involved and a need for research into plant practices.

Related studies of tire manufacturing industry and rubber industry have been published elsewhere. For example, the US Occupational Safety and Health Administration warned of excessive cancer mortality in the rubber industry, including tire manufacturing in 1980 [1]. The International Agency for Research on Cancer also designated "the rubber industry" as carcinogenic (Group 1) because "workers employed in the industry before 1950 have a high risk of bladder cancer, probably associated with exposure to aromatic amines. Leukaemias had been associated with exposure to solvents and with employment in back processing, tire curing, synthetic rubber produc-

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tion, and vulcanization. Excess mortality from lymphomas had been noted among workers exposed to solvents in such departments as footwear and in tire plants. Other cancers, including those of the lung, renal tract, stomach, pancreas, esophagus, liver, skin, colon, larynx, and brain, had been reported as occurring in excess in various product areas and departments." They concluded, however, that no consistent excess of any of these cancers was seen across the various studies [2].

Other studies of cardiovascular disease rates in the rubber/tire manufacturing industry had been undertaken, with inconsistent results. Wingren [3] found an excessive mortality rate among workers who had worked for less than 6 months in Swedish tire manufacturing. The US National Institute for Occupational Safety and Health carried out a case-control study of tire manufacturing, but did not find a link between heat conditions and myocardial infarction [4]. Wilcosky and Tyroler [5] could not prove that organic solvents were the cause of ischemic heart failure in rubber industry workers.

Tire manufacturing and rubber industry environments have been monitored across several European countries. The target substances for this air monitoring were n-nitrosamine, inhalable aerosol, cyclohexane-soluble matter, and specific solvents [6]. Rubber fume occur in the mixing and milling of natural rubber or synthetic polymers combined with chemicals, and in the processes which convert the resulting blends into finished products [7]. No-observed-adverse-effect-level of rubber fumes has not been identified, but the UK assigns maximum exposure limits of rubber fume as  $0.6$  mg/m<sup>3</sup> cyclohexane soluble material [7]. Although a specific carcinogen has not been

**Table 1.** Description of processes in tire manufacturing plants

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clearly identified, rubber fume has been shown to contain small variable amounts of mutagenic substances [8].

The work environments in the tire manufacturing plants are monitored by employers in order to meet Korean regulations every 6 months. The employer in this case study had monitored the total particulates, organic solvent, and noise levels. All of these values were low, compared to the Korean Occupational Exposure Levels.

Several countries (Sweden, UK, Poland, and Germany) initiated prospective cohort studies to address the risk in the contemporary rubber industry. The improved exposure assessment for prospective cohort studies and exposure control in the Rubber Manufacturing Industry was initiated with the European Union [6]. The health risk in the rubber industry is still controversial. An outbreak of sudden cardiac death and cancer cases in a tire manufacturing initiated the concern on the work environment in the Korean tire manufacturing industry for the first time. This epidemiological study was designed at the request of the Ministry of Labor in order to investigate the excessive mortality rate and work environments closely in the tire manufacturing plants. Our purpose is to evaluate the work environment extensively in tire manufacturing plants and to identify process workers' exposure to hazardous substances.

## **Materials and Methods**

### **Study object**

The target tire manufacturing company has two plants (plant A and plant B). This company produces various sizes of pas-



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senger car tire, light truck tire, and bus tire. There are hundreds of tire recipes and several types of tires are produced in workplaces at any one point. The process of tire manufacturing in the plants is described in Table 1.

Sixty-six substances were used to make the tires, either as raw ingredients or as process materials. All substances used by the company are listed in the book "Toxicity and Safe Handling of Rubber Chemicals" [9]. The target substances for air monitoring were determined on the basis of information of this book because it provided information regarding monomer, decomposition substances, and so on. These included aromatic oil extended styrene-butadiene rubber (SBR), which may have high polycyclic aromatic hydrocarbon content [9]. Styrene and 1,3-butadien may also exist as residues or decomposition elements of SBR. Carbon black and calcium carbonate were also used as reinforcing agents and fillers, and n-cyclohexylthio phthalimide was used as retarder. N-nitrosodiphenylamine, a powerful nitrosating agent and may help in producing nnitrosamines, was not used. Sulfur was used as a vulcanizing agent, which suggested that  $SO<sub>2</sub>$  may be released in the air at a high temperature [9]. Aromatic process oil also has a high content of polycyclic aromatic hydrocarbons (PAHs). Another risk is that the bonding agent may decompose into formaldehyde. Aliphatic hydrocarbon solvents are used as the rubber solvent. Carbon disulfide may be produced during the vulcanization process [9]. Furthermore, carbon monoxide may be produced in the workplace because diesel-powered forklifts were used to move rubbers materials and intermediate products.

#### **Methods**

Air monitoring was undertaken on chemicals used in production, the residues of polymer, monomer, and by product. Rubber fume, a cyclohexane soluble material, was chosen for air monitoring. Airborne PAHs, styrene, 1,3-butadien, sulfur dioxide, carbon disulfide, hydrocarbons, and rubber fume were all measured. The bulk rubber solvent was analyzed to identify its





NMAM: National Institute for Occupational Safety and Health Manual of analytical method, OSHA: Occupational Safety and Health Adminstration, KOSHA: Korea Occupational Safety and Health Agency, MDHS: methods for the determination of hazardous substances, PTFE: polytetrafluoroethylene, IOM: Institute of Occupational Medicine, GC: Gas Chromatography, min: minute.

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composition. Table 2 shows our sampling and analytic methods in examining hazardous airborne substances [10-13]. Personal sampling in the workers' breathing zone was conducted during an individual's full shift in the tire manufacturing plants between October and December 2007. Integrated samples were sampled for more than 6 hours. Samples were analyzed in a laboratory which operates a Korean quality control program for industrial hygiene lab and, since 1992, has participated in the Proficiency Analytical Testing program of the American Industrial Hygiene Association.

Carbon monoxide and heat were monitored using direct reading instruments in several spots. Noise was monitored using a noise dosimeter during a full shift on the personal ear zone (Table 2).

Total particulates were measured every 6 months from workers who were handling raw ingredients and rubbers, and these plants had data from 2003 to 2007 (Figs 1, 2). To predict past concentrations of rubber fume, total particulate and rubber fume were sampled simultaneously in the workplaces. After identifying the correlation between the concentration of total particulate and rubber fume, past rubber fume concentrations were predicted with regression analysis that used the total particulate data from 2003 to 2007.

#### **Results**

Nine kinds of PAHs were detected in workers' personal air samples in the tire manufacturing plants. The concentration of PAHs could not be quantified, however, because it was under a limit of quantitation (LOQ). Naphthalene, acenaphthylene, phenanthrene, benzo(B)fluroanthene, benzo(K)fluoranthene, and benzo(G,H,I)perylene were detected in the compounding



**Fig. 1.** Maximum concentration of total particulate in tire manufacturing plant A by year and process. F: first half the year, S: second half of the year.



**Fig. 2.** Maximum concentration of total particulate in tire manufacturing plant B by year and process. F: first half the year, S: second half of the year.





PAHs: polycyclic aromatic hydrocarbons.

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process (Table 3). Naphthalene, Acenaphthylene, phenanthrene, pyrene, benzo(B)fluroanthene, benzo(K)fluoranthene, benzo(G,H,I)perylene, and dibenzo(A,H)anthracene were detected in the curing process. Only naphthalne and acenaphthylene were detected in the extrusion, bead building, calendaring, and tire building processes. The limit of detection (LOD) of Naphthalene, acenaphthylene, phenanthrene, benzo(B)fluroanthene, benzo(K)fluoranthene, and benzo(G,H,I)perylene were 1.00, 0.67, 0.07, 0.36, 0.13 and 0.38 μg/sample, respectively (Table 4).

tected in the compounding and curing process that the rubber fume was evolving. Concentrations of styrene and MIBK were shown in Table 5. The highest concentration of styrene was 0.011 ppm and that of MIBK was 0.722 ppm. Although MIBK was not residue or decomposition, MIBK was detected in all samples.

1,3-Butadiene was not detected in the compounding, extrusion, curing, and production management processes (Table 6). LOD of 1,3-butadiene was 0.15 μg/sample.

The highest concentration of sulfur dioxide was 0.083 ppm in the curing process (Table 7). LOD of sulfur dioxide was



Substances	LOD $(\mu g/sample)$	<b>Substances</b>	LOD $(\mu g/sample)$
Naphthalene	1.00	Benzo(A)anthracene	0.23
Acenaphthylene	0.67	Chrysene	0.19
Acenaphthene	0.92	Benzo(B)fluroanthene	0.36
Fluorene	0.23	Benzo(K)fluoranthene	0.13
Phenanthrene	0.07	Benzo(A)pyrene	0.24
Anthracene	0.06	Indeno(1,2,3-CD)pyrene	0.19
Floranthene	0.21	Dibenzo(A,H)anthracene	0.38
Pyrene	0.11	Benzo(G,H,I)perylene	0.38

**Table 4.** Limit of detection (LOD) values for polycyclic aromatic hydrocarbons

**Table 5.** Airborne concentration of styrene and methyl isobuthy kethone (MIBK) in tire manufacturing plants

	Plant A			Plant B			
	No. of samples	MIBK range, ppm (GM)	Styrene range, ppm	No. of samples	MIBK range, ppm (GM)	Styrene range, ppm	
Compounding		0.722	<b>ND</b>		$0.065 - 0.095(0.078)$	ND-0.011	
Curing		$0.121 - 0.722(0.215)$	<b>ND</b>	3.	$0.068 - 0.339(0.166)$	<b>ND</b>	

GM: geometric mean, ND: not detected.

Limit of detection of styrene: 0.15 μg/sample.

#### **Table 6.** Airborne concentration of 1,3-butadiene in tire manufacturing plants



ND: not detected.

Limit of detection of 1,3-butadiene: 0.53 μg/sample.

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**Table 7.** Airborne concentration of sulfur dioxide in tire manufacturing plant



ND: not detected.

Limit of detection: 1.6 μg/sample.

1.6 μg/sample.

The highest concentration of formaldehyde was 0.029 ppm in the compounding process (Table 8). Formaldehyde was detected in all samples in the compounding and curing processes.

Table 9 and Table 10 show the concentration of hydrocarbons. The concentration of hexane isomers was sum of 2-mehtylpentane and 3-methylpentane because Threshold Limit Values (TLVs) designates hexane isomers, other than

#### **Table 8.** Airborne concentration of formaldehyde in tire manufacturing plant



GM: geometric mean.

#### **Table 9.** Airborne concentration of hydrocarbons in tire manufacturing plant A



Values are presented as range (geometric mean).

ND: not detected.

Limit of detection (μg/sample): cyclopentane (0.13), 2-methylpentane (0.16), 3-mehtylpentane (0.2), n-hexane (0.15), cyclehexane (0.15), 2-mehtylhexane (0.18), 3-mehtylhexane (0.21), n-heptane (0.11), methylcyclohexane (0.11).

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#### **Table 10.** Airborne concentration of hydrocarbons in tire manufacturing plant B



Values are presented as range (geometric mean).

ND: not detected.



**Table 11.** Airborne concentration of carbon disulfide in tire manufacturing plants

ND: not detected.

Limit of detection: 1.2 μg/sample.

n-hexane. The concentration of heptane was the sum of 2-mehtylpentane, 3-methylpentane, and n-heptane because TLVs designates heptane isomers. Six substances detected in the samples were ingredients of bulk rubber solvent used in tire fixing work. When analyzing the bulk rubber solvent, methylcyclohexane content was  $60\%$  (w/w) and n-hexane was  $0.1\%$ (w/w). Benzene was free in bulk rubber solvents. The maximum concentration was found in methylcyclohexane at 3.560 ppm. The concentration of methylcyclohexane in the air was 15 ppm during 20 minutes of tire fixing work.

Carbon disulfide was under the LOQ in compounding, curing, and production management processes (Table 11). LOD of carbon disulfide was 1.2 μg/sample.

Table 12 shows the results for the concentration of rubber fume. Three samples exceeding 0.6 mg/m<sup>3</sup> were found in the curing and production management processes. Geometric mean concentration of the rubber fume was  $0.366$  mg/m<sup>3</sup> in the curing process. The characteristics of aerosol were different between processes because particulate and fume were produced in compounding but only fume was produced in the curing process. The ratio of the concentration of rubber fume to the concentration of particulate in the Institute of Occupational Medicine sampler was different by process. Specifically, the ratio was 22.9 % in compounding and 77.8% in curing.

Noise was a common risk factor through workplaces. Forty-seven percent of workers were exposed to noise exceeding 85 dBA. The results are shown in Table 13.

Heat was measured in December because there were many hot processes in the workplace. The production management workers whose jobs involved changing molds in curing

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<b>Process</b>	No. of samples	Range $(mg/m^3)$	Geometric mean (mg/m <sup>3</sup> )	Ratio $(\%)^*$
Compounding		$0.020 - 0.339$	0.105	22.9
Extrusion	6	0.059-0.166	0.109	44.1
Curing	14	0.212-1.686	0.366	77.8
Production management	16	$0.031 - 1.155$	0.191	56.7
Finishing	2	0.192-0.235	0.212	18.7
Inspection	3	$0.136 - 0.216$	0.161	61.5

**Table 12.** Airborne concentration of rubber fume in tire manufacturing plants

\*Rubber fume concentration to dust concentration in the Institute of Occupational Medicine sampler  $\times$  100.

**Table 13.** Personal noise levels in tire manufacturing plants

<b>Process</b>	<b>Plant A</b>			<b>Plant B</b>			
	No. of samples	No. of samples above 85 dBA	Mean (range), dBA	No. of samples	No. of samples above 85 dBA	Mean (range), dBA	
Compounding	7	5	85.7 (79.0-88.4)	14	8	85.1 (76.1-88.4)	
Extrusion	10	8	85.2 (78.0-87.4)	$\overline{2}$	$\mathbf 0$	82.1 (81.4-82.7)	
Rolling	5	3	85.7 (77.0-89.7)	3	2	83.2 (75.5-85.7)	
Finishing	3	2	86.0 (76.6-90.1)				
Bead building	9	4	84.6 (80.9-90.0)				
Calendering	13	6	84.9 (78.5-89.4)	3	1	82.9 (80.5-86.2)	
Tire building	12	1	82.1 (75.5-86.3)				
Curing	3	1	83.7 (76.0-89.4)				
Production management	3	$\mathbf 0$	83.2 (80.8-84.7)				
Facility maintenance				3	1	84.6 (80.5-89.3)	

**Table 14.** Relative humidity and WBGT in tire manufacturing plants



WBGT: wet bulb globe temperature, PCR: passenger car tire.

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**Fig. 3.** Predictions of past concentrations of rubber fume using the concentration of total particulate.

were exposed to  $28.1^{\circ}$ C (wet bulb globe temperature [WBGT] value) even when the outdoor atmosphere was 2.7°C (WBGT value). The results of temperature are shown in Table 14 and are very different between places. The concentration of carbon monoxide was under 1 ppm through the workplace.

The concentration of total particulate was significantly associated with the concentration of rubber fume in tire manufacturing. The correlation between the concentration total particulate and the rubber fume during the compounding process was high ( $r = 0.85$ ), as was that in curing ( $r = 0.81$ ). A regression equation that could predict the concentration of rubber fume was developed for each process ( $y = 0.44x +$ 0.015 in compounding;  $y = 0.81x + 0.049$  in curing). The past concentrations of rubber fume were predicted using data that had measured total particulate from 2003 to 2007 (Fig. 3). The predicted concentration of rubber fume exceeded the UK regulations on acceptable levels  $(0.6 \text{ mg/m}^3)$  in some samples.

## **Discussion**

In this study, we examined the levels of carbon disulfide, 1,3-butadiene, formaldehyde, styrene, MIBK, sulfur dioxide, and PAHs that are released into the air during the tire manufacturing processes. However, the concentrations in almost all samples were under LOQ or not detected. The concentration of individual chemical was too low or not detected to describe the exposure to hazardous substances in the tire manufacturing plants.

Workers suffered from exposure to rubber fume of more than 0.6 mg/ $m<sup>3</sup>$  in the curing process and in the production management process. Although rubber fume has not been widely studied, Maximum Exposure Limits (MELs) of 0.6

mg/m<sup>3</sup> cyclohexane soluble material do exist [14]. Rubber fume was reported to have mutagenic activity in the mixing and curing process [8] and "company" was identified as the most important exposure determinant for mutagenic activity [15]. This is likely to suggest that the toxicity of rubber fume is different from the company.

The concentration of rubber fumes found during the curing process in this study was higher than that in the new tire production and similar to that of retread tire production found in other countries [16]. The concentration of rubber fume that existed during curing in this study was higher than in the rubber manufacturing in the UK during the 1970s [7]. Furthermore, our predictions of the past concentrations of rubber fume exceeded 0.6 mg/m<sup>3</sup> in both the compounding and curing processes.

The characteristic of total particulate was different by process. Cyclohexane extract existed in 22.9% of inhalable particulate in compounding and 77.8% in the curing process. This suggests that 77.8% of inhalable particulate was cyclohexane soluble material in the curing process. The ratio of cyclohexane soluble matter among particulates was different by process. There was a possibility of misclassification of exposure group if the concentration of total particulate or inhalable particulate is used to classify the exposure group in tire manufacturing.

There is no Occupational Exposure Level for rubber fume in Korea. The work environment in tire manufacturing is deemed to be safe on the basis of the concentration of total particulate. In the past, employers in tire manufacturing plants were not asked to improve the work environment because the concentration of total particulate was under the Korean Occupational Exposure Level. However, there is a need to improve the ventilation system of the curing process on the basis of concentration of rubber fume.

 Forty-seven percent of workers were exposed to noise levels of more than 85 dBA. The physical workload combined with noise or shift work are likely to be responsible for changes to systolic blood pressure, which is a key pathway to coronary heart disease risk [17]. However, noise does not appear to be a direct cause of sudden death because noise is not a specific risk factor in tire manufacturing plants.

Heat stress may be a possible risk factor in the summer. The temperature and humidity in the bead building area is constant for the purpose of production. Likewise, the atmosphere in the curing process areas is hot and humid irrespective of the season. Workers have complained regarding the temperature and humidity in the summer. Although extreme temperature is recognized as a risk factor for coronary heart disease, little is known about the possible cardiovascular effect that tempera-

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ture has among those currently employed by the plant [18].

To better classify exposure groups and to improve work environments, examining closely at rubber fume components and temperature as risk indicators in tire manufacturing is recommended.

## **Confl ict of Interest**

No potential conflict of interest relevant to this article was reported.

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