



Short communication

Decreasing biotoxicity of fume particles produced in welding process

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ABSTRACT

Welding fumes contain heavy metals, such as chromium, manganese, and nickel, which cause respiratory diseases and cancer. In this study, a SiO₂ precursor was evaluated as an additive to the shielding gas in an arc welding process to reduce the biotoxicity caused by welding fume particles. Transmission electron micrographic images show that SiO₂ coats on the surface of welding fume particles and promotes particle agglomeration. Energy dispersive X-ray spectroscopy further shows that the relative amount of silicon in these SiO₂-coated agglomerates is higher than in baseline agglomerates. In addition, *Escherichia coli* (*E. coli*) exposed to different concentrations of pure SiO₂ particles generated from the arc welding process exhibits similar responses, suggesting that SiO₂ does not contribute to welding fume particle toxicity. The trend of *E. coli* growth in different concentrations of baseline welding fume particle shows the most significant inhibition occurs in higher exposure concentrations. The 50% lethal logarithmic concentrations for *E. coli* in arc welding particles of baseline, 2%, and 4.2% SiO₂ precursor additives were 823, 1605, and 1800 mg/L, respectively. Taken together, these results suggest that using SiO₂ precursors as an additive to arc welding shielding gas can effectively reduce the biotoxicity of welding fume.

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1. Introduction

Welding is a process widely used for joining metals. It is estimated that more than 5 million welding workers are exposed to welding fume on a daily basis worldwide [1]. In the United States, approximately 400,000 workers are classified as full-time welding workers [2]. The welding fume forms particles of sizes from the nanometer scale to hundreds of micrometers, with a mode diameter in the interval 0.1–1.0 μm. These welding fume particles have a high probability of being deposited in the alveolar regions of the lungs [1]. The heavy metals present in welding fume—mainly chromium, manganese, nickel, and iron—cause occupational respiratory diseases, including metal fume fever, bronchial asthma, chronic obstructive pulmonary disease, pneumoconiosis, and lung cancer [3]. Thus, issues associated with welding fume for decreasing the risk of welder's health have been intensively investigated in recent years.

Many studies have indicated that the metals that evaporate during the welding process and subsequently form fume particles are the major cause of toxicity to workers [4–6]. For this reason,

finding efficient controls to decrease the toxicity of heavy metals evaporated from the welding process is the most direct way to protect workers. Shielding gas is widely used in welding processes to protect the molten metals within the arc against oxidation and to create favorable arc characteristics [7]. Many scholars have focused on the composition of shielding gas as a tool to decrease the fume formation rate [8]. Although tuning the composition of shielding gas can decrease fume formation rate, it does not significantly affect the fine particle fraction in the welding fume [7,9].

It has been reported that tetraethyloxysilane (TEOS), a silica precursor compound, is useful in controlling heavy metal emission from combustion [10–12]. In the process of gas tungsten arc welding (GTAW), TEOS is also effectively converted into SiO₂ at the high temperatures generated by the welding arc and is able to condense on and coat the surface of the metal particles in the fume. This not only lessens the availability and, hence, toxicity of heavy metals released from welding fume, but also shifts the particle size mode upward by agglomeration and functionally suppresses formation of ultrafine particles [13].

This study aimed to evaluate the biotoxicity of SiO₂-coated welding fume particles. In lieu of the water-sensitive liquid TEOS, stable, volatile tetramethylsilane (TMS) was used as a silica precursor. In our experiments a gas metal arc welding (GMAW) system—the technology most widely used in the weld-

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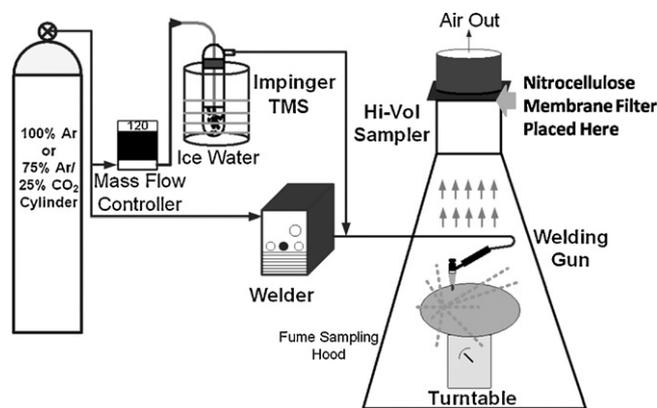


Fig. 1. Welding fume generation and sampling system.

ing industry—generated either common welding fume particles or SiO₂-coated particles. The composition, morphology, coating, and biotoxicity of these particles were characterized and compared.

2. Experimental methods

2.1. Welding fume generation, sampling and characterization

Fig. 1 shows the schematic of the fume generation and sampling system. The chamber used for collecting welding samples was based on ANSI/AWS Method F1.2-1999 [14]. The base metal was placed on a T-25 Aircrafter rotating turntable (MK Products, CA, USA) and the welding torch was fixed in place 2 cm above the turntable. A PowerMIG 140C Gas Metal Arc Welder (Lincoln Electric Company, Cleveland, OH, USA) was used to generate common welding particles and SiO₂-coated particles. In the GMAW process, the electrode is consumed and inevitably produces metal particles. Hence, a Maxstar 150 STL Gas Tungsten Arc Welder (Miller Electric Manufacturing Company, Appleton, WI, USA) that would create a similarly high temperature without creating metal particles was used for generating pure SiO₂ particles. The GMAW process used ER 308L stainless steel welding wire (Harris Corporation, Melbourne, FL, USA), which contains 19.5–22.0% chromium, nickel (9.0–11.0%), manganese (1.0–2.5%), molybdenum (0.75%), copper (0.75%), carbon (0.03%) and silica (0.3–0.65%) to stabilize iron (62.3–68.7%) in the weld.

For GMAW, a cylinder gas of 75% argon and 25% carbon dioxide (Airgas, Radnor, PA, USA) was used as the shielding gas, and a cylinder of ultrahigh-purity argon (Airgas, Radnor, PA, USA) was used as the GTAW shielding gas. In this study, TMS (99.9%, CAS # 75-76-3, Acros Organic, Geel, Belgium) was used as the silica precursor. An impinger containing TMS was set in a water bath at 0 °C to maintain a constant rate of evaporation, and a mass flow controller regulated the proportion of TMS in the shielding gas. A nitrocellulose membrane filter of 90 mm diameter (Millipore, Billerica, MA, USA) and a high-volume pump drawing 1270 Lpm were used to collect particles. Sampling time for each membrane was controlled at 85 s to avoid drastic change in sampling conditions due to dust cake build-up.

The welder was operated at around 11.6 V and the wire feeding speed controller was set to about 1.6 m/min during generation of both common welding fume particles and SiO₂-coated particles. The shielding gas flow rate was 30 Lpm, and 0%, 2%, or 4.2% of the gas passed through the TMS impinger. During generation of SiO₂ particles, the welder was operated at 135 A, the shielding gas flow rate was 7.1 Lpm, and 3% of the feed gas flowed through the TMS impinger.

Transmission Electron Microscopy (TEM; 2010F, JEOL, Boston, MA, USA) was used to visualize the SiO₂ layer coating on the surface of welding fume particles. Energy dispersive X-ray spectroscopy (EDS) was used to compare the composition of baseline and SiO₂-coated welding particle agglomerates. Particles were collected by holding a Lacey Carbon Type-A grid (300 mesh, Pelco, Redding, CA, USA) designed for TEM sampling in front of the high-volume pump for 10 s during the welding process.

2.2. Toxicity test

After sampling, all membranes were dried and weighed, and then the particles on the membranes were washed down with and suspended in sterilized 10 mM phosphate buffer (prepared from 0.18 g/L KH₂PO₄, 1.52 g/L K₂HPO₄ and 8.5 g/L NaCl, and adjusted to pH 7.0). The washed membranes were dried and reweighed, and the weight difference was used to determine particle concentration in the phosphate buffer. In this study, a medium control (0×), the original particle suspension and four dilutions (0.1×, 0.25×, 0.5× and 0.75×) of the particle suspension were used in toxicity tests. The concentration levels of the particle suspension were in the range used in other studies examining nanoparticles toxicity to bacteria (10–5000 mg/L) [15].

Escherichia coli (*E. coli*), which has been widely used for bio- and eco-toxicity testing of particles [15–18], was used in this study. While a mammalian cell line is the best for studying human health effects, the advantages of using *E. coli* bioassay, such as low cost, ease to incubate, fast growth and sensitivity to hazardous materials, make it suitable for initial biotoxicity evaluation. An ampicillin-resistant *E. coli* strain derived from XL1-Blue competent cell (Agilent Technologies, Santa Clara, CA, USA) was used in the biotoxicity tests to avoid background contamination. *E. coli* was grown aerobically at 37 °C overnight in Lysogeny Broth (LB) medium, which contained 10 g/L tryptone, 10 g/L NaCl, 5 g/L yeast extract, and 50 mg/L ampicillin. The saturated cultures were concentrated by centrifugation (Fisher Scientific, Pittsburgh, PA, USA) at 6000 rpm, washed with phosphate buffer, and then resuspended in phosphate buffer to obtain a bio-suspension for which a bacterial titer of 10⁷ to 10⁸ colony-forming units (CFU) per milliliter was measured. A biotoxicity test was conducted by adding 1% by volume of a bio-suspension to a 10-mL portion of particle suspension, agitating with a Wrist Action Shaker (Burrell Scientific, Pittsburgh, PA, USA) at room temperature (20 ± 3 °C) for 24 h, plating on LB medium in a petri dish, and incubation for 24 h at 37 °C. Each biotoxicity test was conducted in triplicate. To prevent changing the particle characteristics from those of the welding fume, particle suspensions were not sterilized, and one sample of the original particle suspension was processed without introduction of bacteria, as a control. The bacterial density after 24-h exposure to the particles was determined by counting CFUs on a petri dish loaded with LB medium containing 50 mg/L ampicillin. Regression analysis was used to determine the LLC₅₀ (the concentration of the welding fume particle that causes 50% fatality in logarithmic concentration of the *E. coli*). One-way analysis of variance (one-way ANOVA) was conducted to differentiate among groups.

3. Results and discussions

3.1. Image of SiO₂-coated welding fume particles

During the GMAW process, the arc temperature directly below the wire was predicted to be higher than 18,000 K, dropping drastically to between 2000 and 5000 K at the edge of the arc [19]. The steep temperature decrease at the edge of the arc causes the metal vapor evaporating from the arc to supersaturate and pro-

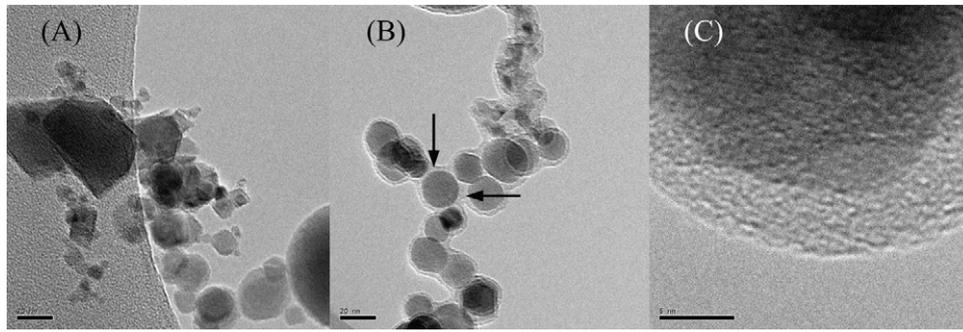


Fig. 2. TEM images: (A) baseline; (B) SiO₂-coated welding particle indicated by arrowheads; and (C) the surface of SiO₂-coated welding particle.

notes nucleation [12]. In this environment, metal particles from nucleation can mix with a silica precursor in the shielding gas at around 1700–1800 K and acquire a coating of SiO₂ on their surface [20]. Fig. 2 shows TEM images of welding particles prepared from this process. In Fig. 2A, the dark spots are welding fume metals that are covered by a very thin layer of SiO₂. This image reveals that the silica contained in the welding wire can form a SiO₂ coating on the surface of the metal particles in arc welding process; however, the amount of silica in the wire was merely 0.3–0.65% and only covered a fraction of the metal particles. Fig. 2B shows an aggregate of welding fume particles formed in the presence of the TMS additive, which deposited a thicker layer of SiO₂ on the surfaces and also as an adhesive between metal particles. Fig. 2C is a magnification of the surface of a SiO₂-coated particle generated by an arc welding process in the presence of added TMS. It reveals that the coating of SiO₂ formed by thermal decomposition of TMS during arc welding is amorphous, and thus has insignificant toxicity to organisms [21,22]. These images suggest that delivering TMS as a silica precursor in the shielding gas for the GMAW process will not only coat the metal particles and aggregates, but also promote aggregation of the metal particles. Topham [23] measured the particle size distribution and reported an increase of mode size from 20 nm for fume particles without SiO₂ precursor to greater than 300 nm for those with 4.2% TMS additive. The observation further evidences the promotion of aggregation. Table 1 compares the compositions of baseline welding particles and of SiO₂-coated welding particles generated from the process with 4.2% TMS additive in shielding gas. The percentage of silicon in agglomerates of SiO₂-coated particles (3.44%) was higher than that in baseline agglomerates (1.92%) shown in Fig. 2. The higher proportion of silicon would be expected to cover the welding particles more completely. It should be noted that particle formation from metal vapors in welding fume is not homogeneous. Hence, the composition of metals in particles does vary significantly from one particle to the other. Since EDS analysis is not a bulk analysis and the results are for those particles shown in Fig. 2, the percentages listed in Table 1 are not the statistical mean values and definitely should not be used to interpret any change in metal composition due to the TMS process.

Table 1
Composition of welding particle agglomerates analyzed by EDS.

Element	Baseline welding particle		SiO ₂ -coated welding particle	
	Weight (%)	Atomic (%)	Weight (%)	Atomic (%)
O	30.62	60.54	19.95	46.05
Si	1.92	2.16	3.44	4.52
Cr	6.51	3.96	15.78	11.21
Mn	2.41	1.39	9.26	6.22
Fe	38.81	21.98	24.05	15.91
Ni	3.35	1.80	1.69	1.06
Cu	16.39	8.16	25.84	15.02
Total	100	100	100	100

3.2. Biototoxicity of welding fume particles

Fig. 3 shows the viable counts of *E. coli* after 24 h exposure to different types and concentrations of welding particles. Viable counts of *E. coli* were similar after exposure to different concentrations of pure SiO₂ particles generated from arc welding process. This result indicates that toxicity of the amorphous silica particles to *E. coli* was insignificant. In contrast, viable counts of *E. coli* after 24 h exposure to the various types of welding particles decreased with an increase in particle concentration. Addition of 2% TMS into the shielding gas decreased the inhibition by approximately half compared to the baseline welding particles, and addition of 4.2% TMS into the shielding gas caused an apparent improvement in survival of only 10% for a doubling of the TMS concentration. As seen in Fig. 4A, the LLC₅₀s were 823, 1605, and 1800 mg/L for baseline, 2%, and 4.2% TMS additive, respectively. One-way ANOVA shows that

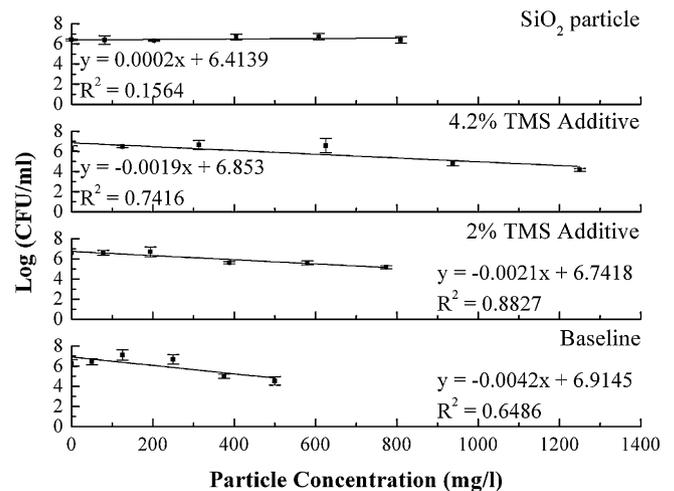


Fig. 3. The growth of *E. coli* after 24 h exposure in different kinds and concentrations of welding particle. The error bars represent one standard deviation.

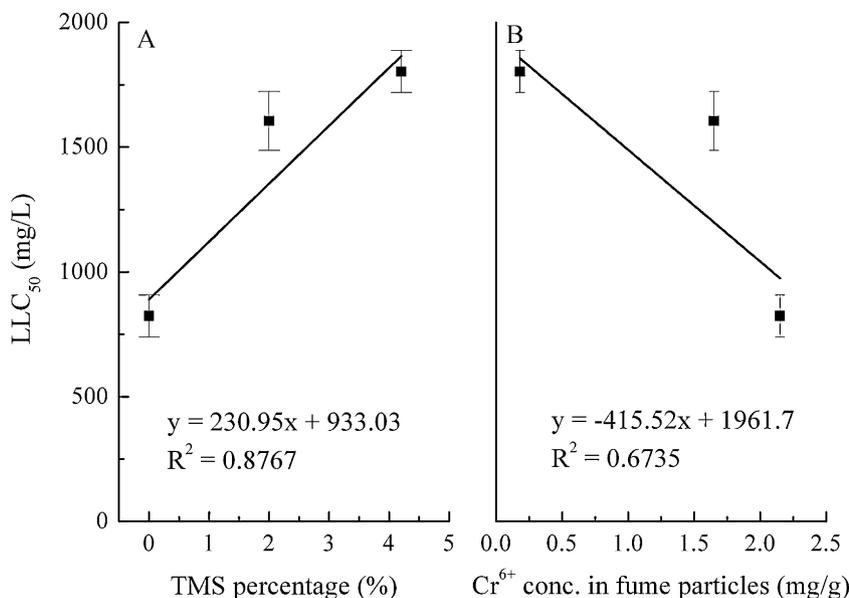


Fig. 4. The correlation of LLC₅₀ to (A) TMS additive fraction; and (B) Cr⁶⁺ concentration in fume particles [22]. The error bars represent one standard deviation.

the difference between treatment groups is significant ($p < 0.005$). These results indicate that adding TMS into the shielding gas could generate fume particles with lower biotoxicity, and further suggest that there might be an optimal concentration of TMS for this purpose, above which the added benefit for increasing the rate of TMS addition falls off.

The LLC₅₀s of both 2% and 4.2% TMS additive in Fig. 4A also show that toxicity persists in SiO₂-coated welding particles. This could arise from incomplete coating of the metal particles with SiO₂ even at 4.2% TMS added. The data of Fig. 4A are insufficient to specify either of these interpretations, but the slight change from 2% to 4.2% TMS suggests the latter is more likely. Topham [23] has reported suppression of Cr⁶⁺ concentration contained in welding fume particles with different concentrations of SiO₂ precursor additive. Fig. 4B shows that Cr⁶⁺ concentration is also significantly correlated with welding fume toxicity. The results suggest that removal of Cr⁶⁺ concentration might be responsible for half of the toxicity while other factors (e.g. insoluble nanoparticles) accounted for the remaining.

4. Conclusions

Using a SiO₂ precursor in the shielding gas of an arc welding process can deposit a SiO₂ coating on the surface of welding fume particles and can also promote agglomeration of the primary fume particles. The toxicity of SiO₂-coated welding fume particles is characterized to be lower than that of the baseline particles. This study suggests that using a SiO₂ precursor in shielding gas is a potentially useful technique for decreasing biotoxicity of welding fume particles. It should be noted that the SiO₂ precursor feeding device has not been optimized for actual work place setting. Furthermore, the influence of the TMS process on welding quality has yet to be verified. Future studies should focus on optimizing operating conditions: SiO₂ precursor, additive volumes, and injection mode in order to achieve the best combination of toxicity, cost, practicability and acceptable welding quality. Meanwhile, toxicity tests as a function of fume particle size distribution and characterization of bioavailability of heavy metals in fume particles are also of importance for clarifying the human impacts of applying a SiO₂ coating to the surface of hazardous welding fume particles. Successful minimization of welding fume biotoxicity allows the SiO₂ precursor

process to be applied to welding processes involving shielding gas, including GMAW and GTAW.

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