Comparison of deposited surface area of airborne ultrafine particles generated from two welding processes

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Abstract
This article describes work performed on the assessment of the levels of airborne ultrafine particles emitted in two welding processes metal-active gas (MAG) of carbon steel and friction-stir welding (FSW) of aluminium in terms of deposited area in alveolar tract of the lung using a nanoparticle surface area monitor analyser. The obtained results showed the dependence from process parameters on emitted ultrafine particles and clearly demonstrated the presence of ultrafine particles, when compared with background levels. The obtained results showed that the process that results on the lower levels of alveolar-deposited surface area is FSW, unlike MAG. Nevertheless, all the tested processes resulted in important doses of ultrafine particles that are to be deposited in the human lung of exposed workers.

Keywords: Welding, ultrafine particles, alveolar-deposited surface area

Introduction
Welding is the principal industrial process used for joining metals and is extensively used in metallic construction worldwide. However, it can produce dangerous fumes that may be hazardous to the welder’s health (Gomes, 1993) and it is estimated that, presently, 1–2% of workers from different professional backgrounds (which accounts for more than 3 million persons) are subjected to welding fume and gas action (Pires et al., 2006). With the advent of new types of welding procedures and consumables, the number of welders exposed to welding fumes is growing constantly, in spite of the mechanization and automation of the processes (Ascenço et al., 2005). Simultaneously, the number of publications on epidemiologic studies (Pires et al., 2006) and the devices for welders’ protection is also increasing. Apart from that, the influence of ultrafine particulate, lying in the nano range, on human health has been pointed to be of much concern (Jenkins & Eager, 2005; Dasch & D’Arcy, 2008, Buonanno et al., 2011), as airborne ultrafine particles can also result from macroscopic common industrial processes such as welding. The detrimental health effects of inhaling ultrafine aerosols were recognised long ago and various attempts have been made to minimize exposure, as the issuing of specific regulations on emissions and objectives for air quality in working microenvironments.

When considering human exposure to airborne pollutants the exposure to airborne particles, and specifically to its finer fractions, such as sub micrometer particles, is of particular interest. In general, the smaller the particles the higher the probability of penetration into deeper parts of the respiratory tract, the exception being made...
to 1–10 nm particles that diffuse very fast and deposit mainly in the nose and mouth. It should be noted that, in air, smaller and larger particles behave differently and, therefore, the penetration of particles of different sizes is also different. Understanding the relationship of airborne nano-sized particulate and human health, under different environmental conditions is of great importance for improving exposure estimates and for developing efficient control strategies to reduce human exposure and health risk and for establishing, evaluating and improving regulations and legislation on air quality. Current workplace exposure limits, that have been established long ago, are based on particle mass. However, this criteria does not seem adequate in what concerns nano-sized particles as these materials are, in fact, characterized by very large surface areas, which has been pointed out as the distinctive characteristic that could even turn out an inert substance into a toxic one, but having the same chemical composition, and exhibiting very different interactions with biological fluids and cells (Oberdörster et al., 1995; Rickerby & Morrison, 2007). As a result, assessing workplace conditions and personal exposure based on the measurement of particle surface area is of increasing interest. It is well known that lung deposition is the most efficient way for airborne particles to enter the body and potentially cause adverse health effects. If ultrafine particles can deposit in the lung and remain there, have an active surface chemistry and interact with the body, then, there is potential for exposure and dosing. Oberdörster (2001) showed that surface area plays an important role in the toxicity of nanoparticles and is the measurement metric that best correlates with particle-induced adverse health effects. Driscoll (1996) considered that the potential for adverse health effects is directly proportional to particle surface area, as confirmed by Card et al. (2008). Surface area per unit of mass of powders is an important product property and the measurement technique used to determine this parameter is the Brunauer, Emmett and Teller (BET) method (Brunauer et al., 1938): the surface area is then determined via the adsorption of nitrogen at the accessible particle surfaces. The major drawbacks are that a large amount of powder is needed, which means that it cannot be applied to airborne ultrafine particles and that it is not an online technique.

It was also noted that mass measurement methods are not sufficiently sensitive for airborne ultrafine particles and are not sensitive toward the specific health relevant properties of the particles (Fissan et al., 2007). The most sensitive particle parameter to measure in relation to health effects of ultrafine particles is the number concentration. It is doubtful that the number concentration correlates well with health effects (Card et al., 2008). This seems to be true for asbestos fibers with a certain probability for each fiber to cause a negative health effect and may be also true for ultrafine particles in case of clogging (agglomeration) after penetrating into the blood. In fact, the health effects after intake are strongly depending also on the deposition regions. Particularly discussed are the deposition in the nose (head), because of possible transfer of ultrafine particles to the brain, the tracheobronchial (TB) region as well as the alveolar region, because of the inefficiency of clearing mechanism and the possible transfer to the blood circulation system with resulting distribution in several end organs (Kreyling et al., 2002).

In 1996, the International Commission of Radiological Protection (ICRP) developed a comprehensive lung deposition model for radioactive aerosols. Several parameters are required to construct the model including breathing rate, lung volume, activity, nose/mouth breathing, etc., and the obtained deposition curves for TB and alveolar deposition) derived from the model can vary according to these parameters. For industrial hygiene applications, American Conference of Governmental Industrial Hygienists (ACGIH) (Phalen, 1999) developed a definition of a reference worker, in order to derive the corresponding deposition curves for TB and alveolar lung deposition, based on the ICRP model: the TB deposition curve represents the fraction of aerosol that deposits in the TB region of the lung, while the alveolar deposition curve represents the fraction of the aerosol that deposits in the alveolar region of the lung. For exposure assessment applications it is common to sample aerosols relevant to their deposition in a specific region of the human lung, which is often referred to as size-selective health hazard sampling. The criterion for size-selective sampling depends on the aerosol being sampled. In what concerns ultrafine particles, the health effects relate to the deposition deep in the alveolar regions of the lung, so the respirable fraction of the aerosol is of interest. The most common online techniques for measuring airborne particle surface area concentrations indirectly are methods based on the attachment of ions or atoms to the particle surface area (Asbach et al., 2009). The epiphaniometer detects, as a measure of relevant surface area, the attachment of lead atoms (210Pb) produced by the decay of Actinium (227Ac) on a particle surface (Gaggeler et al., 1989). This equipment determines the Fuchs (1963) surface area which is proportional to \( \frac{1}{d_p} \) (for \( 30 \text{nm} \leq d_p \leq 150 \text{nm} \)) and therefore represents neither the geometric (proportional to \( d_p^2 \)) nor the lung deposited (more complex \( d_{dep} \) dependency) surface area. Diffusion charger equipments uses an opposed flow unipolar diffusion charger followed by an ion trap to remove excess ions, and the voltage in the ion trap can be adjusted to manipulate the particle size distribution and, therefore, the response function. Downstream of the ion trap, particles are deposited on a filter, and the particle-induced current is measured by an electrometer taken as the measure for lung deposited surface area. Previous tests of this type of equipment demonstrated its reliability in the range of 20–400 nm (Phalen, 1999; Ntziajristos et al., 2007; Kuhlbusch et al., 2000). In spite of certain limitations pointed out by Asbach et al. (2009), with this type of equipment it is possible to determine the surface area concentrations deposited in all regions of the respiratory tract, based on only a single measurement.
by means of calibration factors. Also, if breathing volume and frequency are known, a more health relevant metric, that is, the dose, can be calculated based on the exposure measured with the equipment (Wilson et al., 2007).

**Materials and methods**

For measuring ultrafine particle exposure a nanoparticle surface area monitor, TSI, Model 3550, was used. This equipment indicates the human lung-deposited surface area of particles expressed as square micrometers per cubic centimeter of air (µm²/cm³), corresponding to TB and alveolar (A) regions of the lung. This equipment is based on diffusion charging of sampled particles, followed by detection of the charged aerosol using an electrometer. Using an integral pump, an aerosol sample is drawn into the instrument through a cyclone with a 1 µm cut point. The sample flow is split, with one stream going through a set of carbon and High-Efficiency Particulate Filter (HEPA) filters and an ionizer to introduce positively charged ions into a mixing chamber. The other aerosol flow stream is mixed with the ionized stream in a mixing chamber and charged aerosol and excess ions move onto an ion trap. The ion trap voltage can be set to TB or A response. The ion trap acts as an inlet conditioner or a size-selective sampler for the electrometer, by collecting the excess ions and particles that are not of a charge state, corresponding to the TB or A response settings. The aerosol, then moves on to the electrometer for charge measurement, where current is passed from the particles to a conductive filter and measured by a very sensitive amplifier, as shown schematically in Figure 1. The charge measured by the electrometer is directly proportional to the surface area of the particles passing through the electrometer. The equipment, when set to TB or A response settings, matches the corresponding lung deposition criteria of particles for a reference worker predicted by human lung deposition models from ICRP and ACGIH (Phalen, 1999). In this study, the equipment was set to A response settings as the deposited area in the alveolar tract is the fraction of interest. Particle number concentration and size distribution were measured using a scanning mobility particle size spectrometer (SMPS), TSI, Model 3034. The system consists of three components: (i) a bipolar radioactive charger for charging the particles, (ii) a differential mobility analyzer for classifying particles by electrical mobility, and (iii) a condensation particle counter for detecting particles. The SMPS, TSI, Model 3034, measures the particle diameter (Dp) (in terms of electrical mobility diameter) between 10 and 487 nm using 54 size channels (32 channels per decade) for number concentrations in the range from 102 to 107 #/cm³.

Tests for exposure assessment were made over two different welding processes: metal-active gas (MAG) on carbon steel and friction-stir welding (FSW) of aluminum alloy AA7178, using different welding parameters and also different sampling locations. These welding processes, as well as the base materials used, are among the most used processes in the construction of metallic structures worldwide. Each sampling location and each condition was replicated two times, and a period of stabilization was allowed prior to sampling each condition. The measured deposited area was expressed as alveolar (A) due to the reduced size of the emitted particles. Due to the inexistence of an exposure limit value specific for ultrafine particles, for each measurement task a baseline value was obtained for comparison purposes. MAG welding was performed with a PROMIG Kempi machine using a AWS 70S filler with 0.8 mm diameter of 6 m/min feeding rate under a gas protection of CO₂+15% Ar at 10 L/min flow rate. Three current intensities were tested, 120, 210 and 285 A in order to produce short circuit, globular and spray metal transfer modes. Bead on plate was produced in all cases. All MAG welding operations were performed by the same welder. It should be noted that MAG is a traditional welding process using gases and electric current
in order to provide enough heat to create melting of the added metal wire.

On the other hand FSW is a completely different welding process, which does not require neither gases nor electric current. The welding is produced by the friction strength of a rotating tool being pressed onto two aluminium sheets put together. Figure 2 schematically shows this process. FSW was performed in aluminium alloy AA7178 with a conical threaded probe and a shoulder having a spiral scrolled profile, with a traverse speed (V) of 355 and 180 mm/min and a rotation speed (Ω) of 355 and 1120 rev/mm, that is with V/Ω ratios of 1 and 6, respectively, in hot and cold condition, which means that the conical probe can be heated (in order to facilitate the welding process) or not.

Sampling was taken at different locations inside the welder mask (near the breathing zone) for MAG and at 30 and 60 cm from the weld zone. In FSW, sampling was done close to the shoulder of the tool. The welding masks had no ventilation, and no local ventilation took place during the sampling tests. However, after each sampling procedure took place, the room is ventilated so that the emitted particles do not affect the subsequent measurements.

Results

A summary of the results of measurements made during MAG welding of carbon steel are presented on Table 1, comprising different locations (welder mask and distance from welding front of 30 and 60 cm) and also different operating electrical current intensity ranging from 120 to 285 A. This table also presents values of time weighted average for 8 h of exposure and dose per lung, considering 80 m² of lung area. Figure 3 shows the evolution of measured alveolar-deposited surface area (ADSA) in welder mask, during MAG welding of carbon steel, which took place from 15:50 to 15:51: in this figure, a high ADSA peak of 66,400 µm²/cm² can be noticed, after a gradual increase over the baseline measured at the start-up of the welding task. This peak corresponds to the maximum energy release during welding. After that, a decrease of ADSA can be observed until 15:51 where the welding torch is switched off, corresponding to a decrease of energy intensity released (as this welding process works by electrical pulses). It can be noticed that ADSA still increases from the baseline at beginning of welding. Similar profiles are obtained in measurements done in the welder mask, but using other current intensities. Figure 4 shows the evolution of ADSA during MAG welding of carbon steel, sampled at 60 cm from welding front and using 210 A of electrical current; while Figure 5 shows the evolution noticed at the same sampling location but using an electrical current of 285 A.

The summary of measurements done during FSW of aluminium are presented on Table 2, where two different tool velocities were used, together with two different conditions: cold and hot. Figure 6 shows the evolution of ADSA measured during cold welding at a velocity of 180 mm/min, and Figure 7 shows the evolution of ADSA at the same velocity but during hot welding. Figure 8 shows the evolution of ADSA during cold welding at a velocity of 335 mm/min, and Figure 9 shows the evolution of ADSA at the same velocity but during hot welding. During these four measurements, the measured ADSA evolution profile is similar, but ADSA peaks are higher for lower velocities and for also for hot conditions.

Figures 10 and 11 show, respectively, the measured electrical mobility diameter in MAG welding and in FSW. The obtained average particle diameter and number of particulate are shown in Table 3.

Discussion

In what concerns MAG measurements, and in terms of process parameters, the highest ADSA are obtained for the highest current intensity tested of 285 A, when compared with the lowest intensities of 210 and 120 A. This was to be expected as it was already noticed (Jenkins & Eager, 2005) that the more energy intensive welding processes are, the high amounts of airborne particles are emitted. Pires et al. (2007) studied the metal-transfer mode in MAG welding and the effect of both gas mixture and processing parameters on the fume formation rate. The metal-transfer mode is influenced by the type of the filler wire, voltages and current intensities range, electrode polarity and shielding gas. The arc stability decreases with the increase of CO₂ content in the mixture. This fact is related to the high thermal conductivity of CO₂ which gives rise to more heat loses by conduction and thus the necessity to use higher voltages, for the same current intensity, to initiate and stabilize the arc. The fume formation rate increases with the increase in arc temperature and instability, with the active component, thermal conductivity of the mixture and with the volume of the droplets. The amount of fume released during welding is higher for mixtures with CO₂ relatively to the ones with O₂ having the same oxidizing potential. Differences on the measured values were also observed in relation with the sampling location. As expected the highest values are obtained near the welding front, and as far as the sampling port is from the welding front, lower
values are observed. Values measured inside the welder mask are high which is due to the existence of a confined location as opposed to outside locations where particles dispersion can easily occur. Nevertheless, it should be noted that the particular masks used by welder in this situation are mainly designed to protect the welder from UV radiation and not from airborne emissions.

Measurements were also performed for FSW of aluminum, which has been pointed out as a more cleaner welding process (Nicholas, 1998), as this process does not involve the use of electric current nor the actual fusion and deposition of metal associated with the majority of welding processes. Even for this process, elevated values were measured. As expected, the higher values are obtained for lower velocity of the tool and also for hot condition in comparison with cold condition, as shown on Table 2.

Lee et al. (2007) found numbers of particles per cm³ ranging from 1.69 × 10⁵ to 2.06 × 10⁶ in arc welding performed in conventional booths with a background concentration of 9.08 × 10³, Stephenson et al. (2003) measured around 2 × 10⁵/cm³ during arc welding of carbon steel, and Hovde and Raynor (2007) measured number of particles per cm³ ranging from 9800 using 16 V to 82,800 using 24 V, during arc welding of carbon steel; while Buonanno et al. (2011) measured concentrations ranging from 2 × 10⁵ to 8 × 10⁵/cm³.

Therefore, the number of particles measured in this study is somewhat lower but still of the same magnitude as those measured in other studies.
range of the numbers measured in the mentioned previous studies. It is necessary to point out that these values characterize the exposure at specified distances from sources which tend to be higher, on an order of magnitude, when referring to worker exposure.

It is interesting to note that the measured number of particles emitted is higher in FSW than in MAG welding, which could be due to the fact that MAG welding was done on carbon steel and FSW on aluminium alloys, and emitted steel particles are more dense than emitted aluminium particles, which could mean that steel particles can be easily deposited than aluminium particles and the later could be resuspended. Also the measured particles electric mobility diameter is lower for aluminum particles (around 0.4 times less), which will also contribute to this effect.

Table 2. Measurements for FSW of aluminium.

<table>
<thead>
<tr>
<th>Welding conditions</th>
<th>Sampling location</th>
<th>Sampling time (mm:ss)</th>
<th>Average deposited area (µm²/cm³)</th>
<th>Minimum and maximum values (µm²/cm³)</th>
<th>TWA for 8 h (µm²/cm³)</th>
<th>Total deposited area (µm²)</th>
<th>Dose per lung area (µm²/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No welding: baseline</td>
<td>–</td>
<td>16:00</td>
<td>64 ± 1.2</td>
<td>61.5–68.0</td>
<td>2.11</td>
<td>1.01 × 10⁶</td>
<td>1.27 × 10⁴</td>
</tr>
<tr>
<td>180 mm/min: cold</td>
<td>Welding tool</td>
<td>1:50</td>
<td>10600 ± 180</td>
<td>11.0–42500</td>
<td>40.6</td>
<td>1.95 × 10⁷</td>
<td>2.44 × 10⁵</td>
</tr>
<tr>
<td>355 mm/min: cold</td>
<td>Welding tool</td>
<td>1:20</td>
<td>2500 ± 47</td>
<td>56.0–13900</td>
<td>6.95</td>
<td>3.34 × 10⁶</td>
<td>4.17 × 10⁴</td>
</tr>
<tr>
<td>180 mm/min hot</td>
<td>Welding tool</td>
<td>4:40</td>
<td>16500 ± 285</td>
<td>59.4–100000</td>
<td>160.3</td>
<td>7.70 × 10⁷</td>
<td>9.62 × 10⁵</td>
</tr>
<tr>
<td>355 mm/min hot</td>
<td>Welding tool</td>
<td>3:30</td>
<td>15700 ± 220</td>
<td>38.6–100000</td>
<td>114.5</td>
<td>5.49 × 10⁷</td>
<td>6.87 × 10⁵</td>
</tr>
</tbody>
</table>

cConsidering an average lung area of 80 m² and the following activity related parameters (according to the ICRP/ACGIH model, Phalen, 1999): ventilation rate: 1.3 m³/h; respiratory frequency: 15.0 breaths/min; tidal volume: 1450 cm³; volumetric flow rate: 725 cm³/s.
This set of measurements is the first stage of a study on airborne particles emitted in welding processes. The study clearly demonstrated the existence of ultrafine particles in MAG and in FSW of aluminum, which are clearly dependent from the distance to the welding front and also from the main welding parameters, namely the welding current. The emission of airborne ultrafine particles increase with the current intensity as fume formation rate does. An marked decay of ultrafine particles with the distance to the weld area is observed. When comparing the tested welding processes, FSW process seems “cleaner” than MAG process, as results in lower ADSA. However, the measured number of particles was found to be higher in FSW than in MAG, bearing in mind the different materials used. Nevertheless, it should be noted that FSW has been previously pointed out as a total “clean” welding process as it does not involves direct metal fusion and/or deposition. However, it still produces airborne ultrafine particles and nonneglectable ADSA levels.

It should be noted that, although measured parameters such as the ADSA and the dose per lung area, are elevated when compared with baseline values, they cannot, at this stage, be ascertained as toxicity indicators. Nevertheless, they point out for important contamination of potentially hazardous ultrafine particles released in welding environments.

These preliminary measurements have to be complemented with the size distribution of airborne ultrafine particles and also the chemical composition and information on the shape and crystalline nature of these particles.

**Conclusions**

This set of measurements is the first stage of a study on airborne particles emitted in welding processes. The study clearly demonstrated the existence of ultrafine particles in MAG and in FSW of aluminum, which are clearly dependent from the distance to the welding front and also from the main welding parameters, namely the welding current. The emission of airborne ultrafine particles increase with the current intensity as fume formation rate does. An marked decay of ultrafine particles with the distance to the weld area is observed. When comparing the tested welding processes, FSW process seems “cleaner” than MAG process, as results in lower ADSA. However, the measured number of particles was found to be higher in FSW than in MAG, bearing in mind the different materials used. Nevertheless, it should be noted that FSW has been previously pointed out as a total “clean” welding process as it does not involves direct metal fusion and/or deposition. However, it still produces airborne ultrafine particles and nonneglectable ADSA levels.

**Acknowledgements**

The authors acknowledge Mr. Campos, Mr. Carlos Guerreiro, Mr Daniel Lavourador and Ms. Rita Santos for their assistance during welding tests.

**Declaration of interest**

The authors state no declarations of interest.

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