Review

Ultrafine Particles Emitted by Flame and Electric Arc Guns for Thermal Spraying of Metals

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The ultrafine aerosol emitted by thermal spraying of metals using flame and electric arc processes has been characterized in terms of particle size distribution and emission rates based on both particle number and mass. Thermal spraying of Zn, Zn/Al, and Al was studied. Measurements taken using an electrical low pressure impactor and a condensation nucleus counter reveal an aerosol made up of very fine particles (80–95% of number distribution <100 nm). Ultrafine particle emission rates produced by the electric arc process are very high, the largest values being recorded during spraying of pure aluminium. This process generates high particle emissions and therefore requires careful consideration and possible rethinking of currently implemented protection measures: ventilated cabins, dust collectors, and personal protective equipment.

Keywords: cabin; filtration; thermal spraying; ultrafine particles

INTRODUCTION

Thermal spraying is a surface treatment process, which enables different types of material to be deposited on various substrates: metals, metal alloys, ceramics, and even plastics. The process involves spraying a metal coating product, in wire or powder form, which is melted by a heat source and then sprayed by compressed air onto a surface previously prepared by sandblasting.

Different heating systems can be used to produce molten metal feedstock; the two most common processes to achieve such coating are the flame spray gun and the electric arc spray gun. The molten metal is propelled by combustion gases in the case of flame spraying and by compressed gas (air, nitrogen, and argon) input in the case of electric arc spraying. In all metal spraying processes, large quantities of fumes composed of fine particles are generated, which may present a serious risk to the operator.

The ultrafine nature of the generated aerosol particles has the effect of reinforcing its toxicity (Karlsson et al., 2009).

The metals most commonly used for anti-corrosion surface treatments are zinc, tin, zinc/aluminium alloys, and special alloys sometimes containing highly toxic metals, such as chromium, cobalt, nickel, etc. (Ducos, 1999; Chadwick et al., 1997).

Metal thermal spraying processes are subject to a number of risks, including chemical, due to toxicity of the generated particles and gases, noise-related, ultraviolet and infrared radiation, electrical, fire/explosion, etc. (Howes, 2001). These multiple risks make it essential for these processes to be implemented in a closed ventilated cabin and the operator must be fitted with respiratory protective equipment, typically a compressed air-supplied hood (Guide pratique de ventilation n°14, 2004). The polluted air extracted from the cabin is filtered before being discharged into the atmosphere outside the building. Frequently encountered dust collectors incorporate cartridge filters, which are unlogged by injection of a compressed air counterflow.
In cooperation with the Caisse Régionale d’Assurance Maladie (CRAM) (French regional health insurance fund) for the Rhône-Alpes region, Institut National de Recherches et Sécurité has conducted a study aimed at characterizing metal fumes emitted by both flame and electric arc spraying processes in order to determine possible causes of filter clogging in dust collectors connected to ventilated cabins. Clogging is reflected by reduced ventilation flow rates and hence by increased pollutant concentrations inside these cabins.

EQUIPMENT AND METHODS

Figure 1 illustrates the experimental set-up for measurement of generated aerosol. The pollutant emission generated by the metal coating process is collected by a semicircular suction panel connected to a 35-cm diameter and 1.50-m long duct. The system is placed inside a 63-m³ ventilated cabin fitted with two filtered fresh air blowing outlets located on the back of the collection unit. A diaphragm is positioned at the collection unit outlet to generate turbulence and thereby ensure greater concentration uniformity at the measuring point in the duct.

Probes are arranged inside the duct for sampling the aerosol and enabling it to be analysed using different systems:
- A tapered element oscillating microbalance (TEOM—rp 1400 A) for measuring particle mass concentration, with a sampling airflow rate of 1 l min⁻¹.
- An electrical low pressure impactor (ELPI—Dekati) for measuring particle size distribution for aerodynamic diameters (dae) between 28 nm and 9.9 μm, at an airflow rate of 9.8 l min⁻¹.
- A condensation nuclei counter (CNC—Grimm 5.400) for measuring number concentration for particle diameters >5 nm, at an airflow rate of 1.5 l min⁻¹.
- Sampling on microporous membranes (pore diameter 80 nm and filter diameter 37 mm) previously carbon coated for analysis using a scanning electron microscope (SEM).
- A pitot tube for measuring air velocity.

The 35-mm long and 10-mm internal diameter stainless steel sampling probes are arranged inside the duct at a spacing of a few centimetres to prevent possible interference.

A 1:10 dilution stage (Palas VKL 10) is fitted upstream of the ELPI impactor reducing the sampling airflow rate at 4 l min⁻¹. The same probe is used for CNC counter and TEOM sampling. Both these instruments are connected to the probe through a (TSI) flow splitter. A 1:100 dilution stage (Grimm

![Fig. 1. Experimental set-up for aerosol emission measurement placed inside the ventilated cabin.](https://academic.oup.com/annweh/article-abstract/54/6/607/152066/1)}
Tests 1

The purpose of these tests was to determine the aerosol particle size distribution and the number and mass emission rates generated by arc and flame spray processes. These tests were conducted using a flame (Master Jet Marin) and an electric arc (Margarido M45) spray guns fed with Zn/Al (85/15%) wire feedstock. The wire (diameter \(Df = 3\) mm) feed rate was set to 2.35 m min\(^{-1}\) for the flame spray gun, ensuring a mass feed rate of 6.4 kg h\(^{-1}\). A 180 A current (voltage 23 V) was applied to the electric arc spray gun, fed by a 2.3-mm diameter wire, ensuring a mass feed rate of 14.2 kg h\(^{-1}\). Spraying was directed onto a previously sandblasted flat metal plate positioned in front of the collection unit.

Tests 2

The purpose of these tests was to study the influence of a number of parameters on particle emission from an electric arc spray gun (Margarido M45). These parameters include compressed air pressure, current, wire type and composition, etc. This time metal coating was performed on a complex geometry workpiece comprising a chair leg. Measurements taken only using the TEOM enabled us to determine the aerosol mass emission rate.

Measurements on a dust collector

Measurements were taken with the aim of characterizing the aerosol upstream and downstream of the dust collector, which was installed on a cabin used for electric arc metal coating operations. Testing conditions were a Margarido M45 spray gun fed by Zn/Al (85/15%) wire with a diameter \(Df = 2\) mm, an American Air Filter dust collector incorporating 36 cartridge filters providing a combined filtration area of 920 m\(^2\) (80% cellulose and 20% polyester medium), and unclogging by injection of a compressed air counterflow.

Measurements were taken using an ELPI impactor and by sampling on filters (polyvinyl chloride membranes, Pall GL/A, \(\Phi = 47\) mm) previously weighed using a Mettler AT21 balance. The aerosol was isokinetically sampled by metal elbow probes (internal diameter 16 mm) introduced into the duct. ELPI impactor downstream sampling required inclusion of an additional fitting comprising a carbon-coated antistatic flexible tube (TSI, diameter 10 mm and length = 50 cm). No ELPI impactor measurements could be taken upstream of the dust collector because of the difficulty in reaching the high level (4–5 m) measuring point. The aerosol particle size distribution was therefore characterized inside the cabin. The sampling point was located in the centre of the cabin at a height of 1.50 m; it was connected by a 2-m long antistatic tube to the ELPI impactor placed outside the cabin.

RESULTS AND DISCUSSION

Tests 1

Table 1 presents the main measurement results. The values given in this table are averages of three consecutive test results. The collected airflow was 4860 m\(^3\) h\(^{-1}\).

\(Cn\) and \(Cm\) designate the particle number and mass concentrations and \(\varphi n\) and \(\varphi m\) are the associated particle number and mass emission rates, respectively. \(\varphi ml\) is the mass emission rate of metal consumed. NMD and MMD designate the number and mass median diameters (NMD and MMD) derived from the ELPI impactor data. A particle density of 4.0 g cm\(^{-3}\) was taken for impactor data computation purposes. This density corresponds to the effective density (or apparent density) of the particle \((\rho p_{eff})\). It can be

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Arc plate</th>
<th>Flame plate</th>
<th>Rf arc/flame</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Cn) (ELPI # cm(^{-3}), (dp \geq 28) nm)</td>
<td>(1.6 \times 10^8)</td>
<td>(3.6 \times 10^6)</td>
<td>/</td>
</tr>
<tr>
<td>(\varphi n) (ELPI), (# s(^{-1}) \times 10(^{15}))</td>
<td>216</td>
<td>0.49</td>
<td>441</td>
</tr>
<tr>
<td>(Cn) (CNC # cm(^{-3}), (dp \geq 5) nm)</td>
<td>(1.93 \times 10^9)</td>
<td>(3.1 \times 10^7)</td>
<td>/</td>
</tr>
<tr>
<td>(\varphi n) (CNC), (# s(^{-1}) \times 10(^{13}))</td>
<td>260.4</td>
<td>4.2</td>
<td>62</td>
</tr>
<tr>
<td>(Cm) (TEOM), (mg m(^{-3}))</td>
<td>132</td>
<td>25</td>
<td>/</td>
</tr>
<tr>
<td>(\varphi m) (TEOM), (mg s(^{-1}))</td>
<td>178.2</td>
<td>33.7</td>
<td>5.3</td>
</tr>
<tr>
<td>(\varphi ml), metal (g s(^{-1}))</td>
<td>3.94</td>
<td>1.77</td>
<td>2.22</td>
</tr>
<tr>
<td>NMD (ELPI), (nm)</td>
<td>75.6</td>
<td>50.2</td>
<td>/</td>
</tr>
<tr>
<td>MMD (ELPI), ((\mu m))</td>
<td>0.17</td>
<td>3.2</td>
<td>/</td>
</tr>
</tbody>
</table>

\(Cn\) and \(Cm\): particle number and mass concentrations; \(\varphi n\) and \(\varphi m\): particle number and mass emission rates; \(\varphi ml\): mass emission rate of metal consumed; NMD and MMD: number and mass median diameters of the particle size distribution.
determined from the bulk material density ($\rho_p = 5.73$ g cm$^{-3}$ for that alloy composed of 85% Zn and 15 % Al) and from the particle dynamic shape factor $\chi$ (DeCarlo et al., 2004). From the works of Hand et al. (2002), the effective density can be simply determined from the following relation:

$$\rho_{p_{\text{eff}}} = \frac{\rho_p}{\chi}. \quad (1)$$

The dynamic shape factor can be estimated directly from the number of primary particles, $n_{pp}$, that compose the aggregate (Baron et al., 2001):

$$\chi = n_{pp}^{1/9}, \text{ with } n_{pp} \leq 60. \quad (2)$$

The number of primary particles is most of the time an unknown parameter. But considering the relation 2 and the exponent 1/9, we see that $\chi$ varies not much, between 1.30 ($n_{pp} = 10$) and 1.57 ($n_{pp} = 60$). We take the average value of 1.43 for the determination of $\rho_{p_{\text{eff}}}$. But theories, which describe how varies the dynamic shape factor of a fractal aggregate with the aggregate size give rather divergent results (Baron et al., 2001). So there is a great uncertainty on the determination of the apparent density of an aggregated particle.

The number concentration of the aerosol generated by arc spraying measured with the CNC is slightly above the detection limit of that instrument, fixed at $10^7$ cm$^{-3}$. The CNC concentration is therefore underestimated in this case despite the 1:100 dilution stage.

**Observation 1.** Particle emission rates emitted by the electric arc process are significantly higher than those emitted by the flame process. Electric arc/flame emission rate ratios $R_f$ are also much higher, when expressed in number of particles rather than in mass. This can be explained by the very marked discrepancy between the arc (MMD = 0.17 $\mu$m) and flame (MMD = 3.2 $\mu$m) mass distributions. The number distribution for the ‘flame’ aerosol (Fig. 2) reflects particles of aerodynamic diameter $>1 \mu$m, which become predominant in terms of mass. These particles $>1 \mu$m were not found in the electric arc-generated aerosol within the scope of these tests (Fig. 3).

The observed difference between particle number $R_f$ ratios derived from ELPI and CNC measurements is due to CNC’s detection limit (5 nm) being lower than that of the ELPI (28 nm). However, the number distribution obtained with the flame process reveals a significantly lower NMD than that of the distribution derived from the electric arc measurements (Figs. 2–4). The number concentration of flame particles measured with the ELPI is therefore underestimated, which leads to an artificial increase in the $R_f$ ratio determined using this instrument.
Mass distributions computed from number distributions must often be treated with caution as they tend to over-evaluate the fraction of largest particles (Gulijk et al., 2004), but they clearly explain the recorded differences between the mass and number emission rates in this case.

Finally, the Table 1 data (\(\varphi_m\) and \(\varphi_m^c\)) show that 2–4.5% of the consumed metal in both flame and electric arc processes effectively causes aerosol (fume) formation. The mass of the generated particles could be increased by their instantaneous oxidation (especially for \(\text{Al}_2\text{O}_3\)). Due to this phenomenon, the mass emission rate of these oxidized particles (\(\varphi_m\)) is slightly increased in comparison with the consumed (non-oxidized) metal mass emission rate (\(\varphi_m^c\)). A more detailed mass balance was performed by weighing the plates before and after spraying to determine the metal mass deposited on the target. These measurements show that \(~50\%\) of the sprayed metal mass is effectively deposited on the plate, while the remainder (45.5–48%), comprising metal particles not retained by the target, is found on the cabin walls in the form of sometimes considerable deposits, if there is no regular cleaning. The particle emission converted into the same quantity of consumed metal show that electric arc spraying is by far the most emissive process: 28 times the flame process emissiveness in number (CNC) and 2.4 times in mass (TEOM).

Observation 2. Particles composing fumes are very small with 80–95% of the particle number distribution measured using the ELPI <100 nm. Concentrations are very high, especially for the electric arc spraying process. At these concentration levels (>\(10^9\) # cm\(^{-3}\)), the aerosol is very unstable because of particle coagulation, which clearly complicates its metrology. Considering the dimensions of the sampling probe and the airflow rate of 1.5 l min\(^{-1}\) (CNC), the residence time of the aerosol inside the probe, i.e. between the sampling point and the dilution stage, is of the order of 1 s. This time is long enough to modify both the particle concentration (halved in \(<1\) s) and the particle size distribution (Hinds, 1999). The original aerosol is therefore more concentrated (in number) and probably presents a particle size distribution shifted to smaller particles. This phenomenon could also explain the shift in the arc spraying aerosol particle size distribution towards the larger particle diameters, an effect that is not observed for the flame spraying aerosol (Figs. 2 and 3).

The SEM photograph (Fig. 5) taken using a sample of particles collected on a microporous membrane confirms these observations. It shows that both very small size particles (\(<100\) nm) and micron size agglomerates.

Tests 2

Figure 6 shows the mass emission rates measured with respect to various parameters. The reference situation corresponds to the following configuration: spraying onto tubular target, Zn wire with a diameter \(D_f = 2.3\) mm, spray gun—target distance \(d = 15\) cm, current \(I = 100\) A, and compressed air pressure = 6 bar. The emission rates are plotted with respect to the parameter adjusted against these reference conditions (the other parameters remaining unchanged).

We note significant emission variation with the current, the wire diameter, and the target geometry. The highest emission, which was recorded when spraying onto a flat plate and was otherwise confirmed by other series of measurements, is rather surprising and unexplained.

Spraying a Zn/Al alloy leads to significantly higher emission rates than spraying pure Zn. However, the greatest emission is observed for spraying pure Al, which gives emission rates between 500 and 850 mg s\(^{-1}\).

These specific particle emission rates allow us to predict the concentrations to which a ventilation system will be subjected. For example, an electric arc thermal spraying operation (using data from Table 1) will generate a concentration of \(4.7 \times 10^8\) particles of diameter \(\geq 5\) nm cm\(^{-3}\) in a 20 000 m\(^3\) h\(^{-1}\) ventilation system. This concentration is very much higher than the ultrafine particle (diameter <100 nm) concentrations usually encountered in industrial environments (Evans et al., 2008; Thomassen et al., 2006), namely between \(4 \times 10^4\) and \(2 \times 10^6\) # cm\(^{-3}\).
We see that electric arc spray guns are extremely emissive and effectively generate very high particle concentrations in the vicinity of the source. Mass emission rates recorded for electric arc spraying are, for example, 10–50 times higher than the fume emission rates generated by electric arc welding operations (Mayer et al., 1980). Expressed in terms of number of particles, an arc spray gun emission rate is ~1000 times higher than that generated by an arc welding process (Zimmer and Biswas, 2001).

Site measurements

The cabin ventilation airflow rate and thus that of the dust collector was 15 830 m$^3$ h$^{-1}$. Filters were cleaned by injection of brief pulses of compressed air (technique of pulse-jet cleaning) when a 1820 Pa pressure loss is reached. Recorded mass concentrations were between 52 and 195 $\mu$g m$^{-3}$ downstream and 128 mg m$^{-3}$ upstream. The photograph (Fig. 7) shows particles collected on the upstream sampling filter. The particles composing the fume can be seen around the filter periphery and are clearly distinguishable from the large diameter metal particles (>10 $\mu$m) occupying the central part of the filter.

Figure 8 illustrates the number distribution for the aerosol downstream of the dust collector measured using the ELPI impactor. The average concentration is $5 \times 10^5$ # cm$^{-3}$ with 90% of the number distribution concerning particles with aerodynamic diameters <100 nm.

Although pollutants are not directly comparable, the emission limit for carbon particles from diesel vehicles is shown in the figure. The emissions are calculated per bar of pressure, Amp, and diameter of the spray gun.

Fig. 6. Mass emission rates of aerosol generated by electric arc spray gun for different test configurations—Tests 2.

Fig. 7. Photograph of upstream sampling filter showing metal particle at centre and fume deposit around periphery (orange area).

Fig. 8. Number particle size distribution of aerosol measured downstream of dust collector (ELPI data)—broken/dashed curve: cumulative distribution.
engines of $10^{12}$ particles $>23$ nm kWh$^{-1}$ (Opair, 2008) corresponds to an equivalent concentration limit of $1.7 \times 10^7$ particles $>23$ nm cm$^{-3}$, which is lower than the values recorded downstream of the installation.

The ratio between number particle concentrations measured upstream and downstream the dust collector (ELPI data) allows us to deduce the filtration efficiency. The number efficiency determined in this way exceeds 99.8%.

Measurements taken inside the cabin using the ELPI impactor give particle size distributions in agreement with those recorded during laboratory studies. Figure 9 shows the variation in particle number concentration in the cabin with time after thermal spraying has been stopped. It can be seen that the concentrations reached are especially high ($>10^8$ cm$^{-3}$). The relatively slow decrease in concentration (Fig. 9) agrees with the cabin ventilation data, namely a ventilation time constant $t_c$, defined by the ratio $V/Q$ ($V =$ cabin volume $= 222.7$ m$^3$ and $Q =$ flow), of 50 s. The kinetics of the concentration decrease in a volume under ideal mixing is governed by an exponential law (Fig. 9, dotted curve). The concentration reached at time $t$ is $4 \times t_c^{-1}$, i.e. 3 min 25 s, is in theory $\sim$2% of the initial concentration ($C_n = 1.5 \times 10^8$ # cm$^{-3}$, i.e. $C_n = 3 \times 10^8$ # cm$^{-3}$), which is roughly observed. We can see that the cabin decontamination time is quite long because of the particularly high concentrations. Wearing of this equipment inside the cabin, even after the spraying operation has been stopped, would also appear necessary because of the relatively slow decontamination kinetics. In the present case, a period of $\sim$6 min would be theoretically necessary to reach the industrial hall ambient concentration of $1.5 \times 10^5$ # cm$^{-3}$ (dae $\geq 28$ nm).

**CONCLUSIONS**

The conducted study enabled us to characterize the aerosol generated by electric arc and flame thermal spraying processes. The measurements revealed a totally sub-micron aerosol with an 80–95% ultrafine particle (diameter <100 nm) fraction in numerical terms. Very high emission rates were recorded for electric arc spraying, in particular using pure aluminium, and these values are far higher than those encountered in the arc welding field, for example. Concentrations $>10^8$ particles (diameter $\geq 28$ nm) cm$^{-3}$ were recorded inside ventilated cabins, in which electric arc spraying operations are performed.

At these high particle number concentrations ($10^8-10^9$ cm$^{-3}$), the aerosol is highly unstable due to coagulation. The aerosol characteristics (particle size distribution, number concentration, chemical composition, particle morphology, etc.) can quickly change during the aerosol transport from the sampling point to the measuring device. The characteristics of the original unsampled aerosol are therefore very difficult to assess.

The high particle emissions generated by this process mean that careful thought must be given to the protection measures implemented: ventilated cabins, dust collectors, and personal respiratory protective equipment. Ventilation flows require re-examination, based on the measured emissions, to reduce concentrations inside cabins and decontamination times after stopping a metal coating operation. Similarly, dust collector filter clogging is a common phenomenon in electric arc metal spraying. This sometimes rapid clogging of filters degrades cabin ventilation by sharply reducing the ventilation flow. By increasing filtration areas, in particular, solutions provided by manufacturers can effectively improve the situation but do not really solve the problem. Studies should therefore be conducted to understand filter clogging mechanisms and determine which parameters could be worked on to curtail this phenomenon. The protection efficiency of compressed air-supplied hoods in environments so highly loaded in ultrafine particles should also be verified. Finally, biometrological studies would be particularly instructive, based on the state of surface contamination (cabin, industrial hall, etc.) by a specific deposit.

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