Quench rate affects oxidation reactions in the formation of arc welding fumes

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Abstract: The formation process of fumes in gas metal arc welding (GMAW) has been modelled considering the iron oxidation reactions both in gaseous and condensed phases. The results show that fumes are composed of aggregates of primary particles that are nucleated from gas-phase FeO and further oxidized to Fe_3O_4 or Fe_2O_3, depending on the quench rate. Increasing the latter results in two effects: on one side, it induces the formation of smaller primary particles, which are more easily oxidised, but at the same time it reduces the time for solid-state diffusion of oxygen, resulting in lower particle oxidation.

Keywords: Nanoparticles synthesis, welding fumes, modelling, meso-scale model.

1. Introduction

Fumes formation is a typical side effect of arc welding process and it is widely recognized as an important occupational problem [1]. The hazard level of such fumes depends on their characteristics, such as size and composition, which are strongly dependent on the welding process type and operating conditions [2]. For this reason, the understanding of the formation mechanisms and the possibility to predict and control their characteristics could be an important mean for the hazard reduction.

Gas Metal Arc Welding (GMAW) is one of the most broadly adopted processes in industry as it can be automated and it allows a high deposition rate. Unfortunately, this process is characterized by a strong production of fumes, mainly in the form of aggregates of nanoparticle oxides. Literature have shown that most of the particle fumes are generated from the metal vapours coming from the overheated liquid portion of the wire tip that are transported by convection and diffusion to the arc fringes, where the temperature drops to 1500-2500 K, leading to nanoparticle formation by nucleation from a supersaturated gas. Then nanoparticles continue to grow by surface condensation, coalescence and finally forming aggregates, whose fractal dimension depends on the collision rates and gas phase regime [3].

Chemical analyses of the fumes produced in GMAW of mild steels have shown that nanoparticles are mainly composed of Fe_3O_4 and, in a smaller amount, of manganese oxides. However, a complete understanding of the mechanism of fume formation in oxidant mixtures is missing.

A model for simulating the fume formation mechanism that includes the effects of iron oxidation reactions and enables the prediction of nanoparticle dimensions and morphology, considering also aggregation and coagulation phenomena has been developed by one of the Authors in the past [4]. This model is based on the stochastic approach originally developed for the simulation of nanoparticle synthesis in flame processes in [5]. Simulations of the fume formation process in arc welding are carried out considering the typical temperature profiles and mixture compositions that characterize a GMAW process.

2. Modelling approach

The modelling approach adopted in the present work is based on a stochastic algorithm developed for the analysis of the synthesis of nanoparticles [5]. This model has been extended in [4] to include the physical and chemical phenomena that describe fume formation in thermal plasmas.

The generation of nanoparticles is simulated tracking the temporal evolution of a control volume V_{ctr}, which is assumed to move with the flow along a streamline starting from a high-temperature region (at 3000 K) and ending in the colder region surrounding the plasma (at 300 K). Primary particles formation by nucleation of vapours in the gaseous phase is predicted, calculating the homogeneous nucleation rate as done in [6]. Primary particles growth is predicted by balancing the surface condensation of vapor against evaporation using the approach proposed in [3].

A free molecular collisional kernel is used to predict primary particles coagulation to form aggregates. A majorant kernel approach is used for computational effort reduction, as proposed in [5]. A ballistic algorithm is applied upon collision, to simulate aggregates coagulation and predicts the fractal dimension of the resulting merged aggregate [7]. Sintering and coalescence are considered, using the Friedlander-Koch relation for the sintering time [8]. Intra particle chemical reactions have been modelled using the approach depicted in [4], considering oxidation reactions between particles and the gas-phase.

The thermo-fluid-dynamic simulation of the arc is not considered in this work. The composition of the gas-phase is calculated assuming chemical equilibrium for given temperature, pressure and atom concentration conditions.

The atom concentration in the gaseous phase is tracked by subtracting the vapour converted to nanoparticles. The system is kept at constant pressure whereas the temporal evolution of the temperature is imposed according to a linear profile.

A cooling rate of 10^5 to 10^7 K/s is estimated according to the simulation results reported in [9] for arc welding and nanoparticle synthesis. These results also show that partial pressure of iron vapours in the arc region at 3000 K during GMAW welding can range from 1 to several thousand Pa. In this work, initial iron vapour pressure has been set to 3000 Pa. The oxygen partial pressure has been set to 9000 Pa, which is the maximum amount of oxygen in active mixtures usually adopted in GMAW.

3. Results

Gas and particle phase compositions as function of temperature for two different quenching rates of -10^7 K/s and $-5 \cdot 10^5$ K/s are shown in Fig. 1.

The most important effect of quenching is its influence on the final Fe_3O_4 to Fe_2O_3 ratio in the particle phase. The conversion of Fe_3O_4 into Fe_2O_3, that occurs at temperature around 1500K, is limited below 1400 K by the solid diffusion, that prevents further oxidation of the particles. For this reason, rapid quenching leads to faster solidification and then to a reduction of the final O/Fe ratio.

Composition of the solid phase against cooling rate is plotted in Fig. 2, showing that between 10^5 to 10^7 K/s a non-linear correlation between cooling and oxidation rate can be found, with a minimum on the Fe_3O_4 concentration at about 5 10^5 K/s. Increasing the quench rate results in two effects: on one side it induces the formation of smaller primary particles, which are more easily oxidised, but at the same time it reduces the time for solid-state diffusion of oxygen, resulting in lower particle oxidation.

Particles size distribution for the same abovementioned quenching rates are shown in Fig. 3, comparing the diameter d for the spherical primary particles p_i and the collision diameter d_{col} aggregates P_i. The case with the faster -10^7 K/s quench leads to the formation of a higher number of both primary particles and aggregates with mean diameters of 12nm and 41nm respectively. The case with the lower quench rate of $-5 \cdot 10^5$ K/s leads to a reduction of the number of particles and aggregates, while an increase of the mean diameters for both primary particles and aggregates up to 48nm and 115nm respectively, is achieved. These behaviour is due to the influence of the quenching rate on the time length of the coalescence regime, where primary particles increase their diameter by instantaneously coalescence upon collision. These phenomena are well depicted in Fig. 4, where the actual shapes and morphology of sample aggregates are represented for the two different quenching rates considered above, showing the differences between primary particles diameters and aggregates morphologies as predicted by the model.

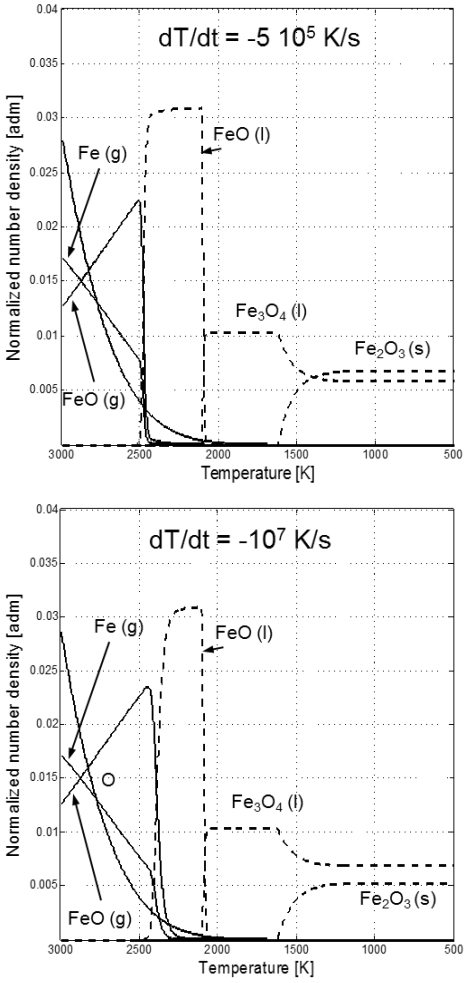

Figure 1 – Gas- and solid-phase composition for two different quenching rates.

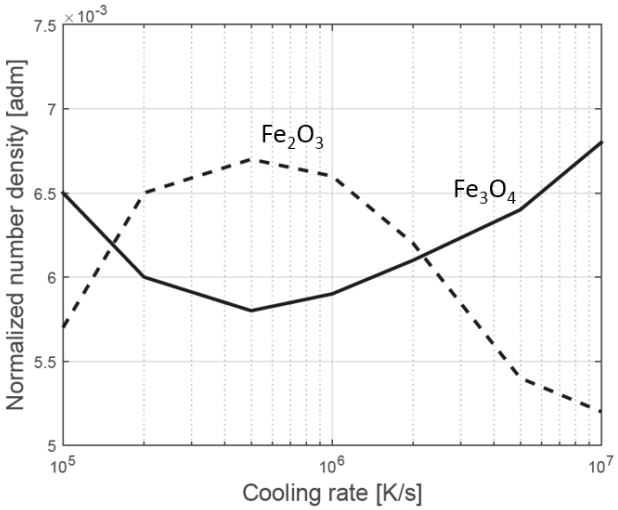

Figure 2 – Solid-phase composition of the particle ensembre at the end of the fume formation process (T = 300 K) as function of cooling rate.

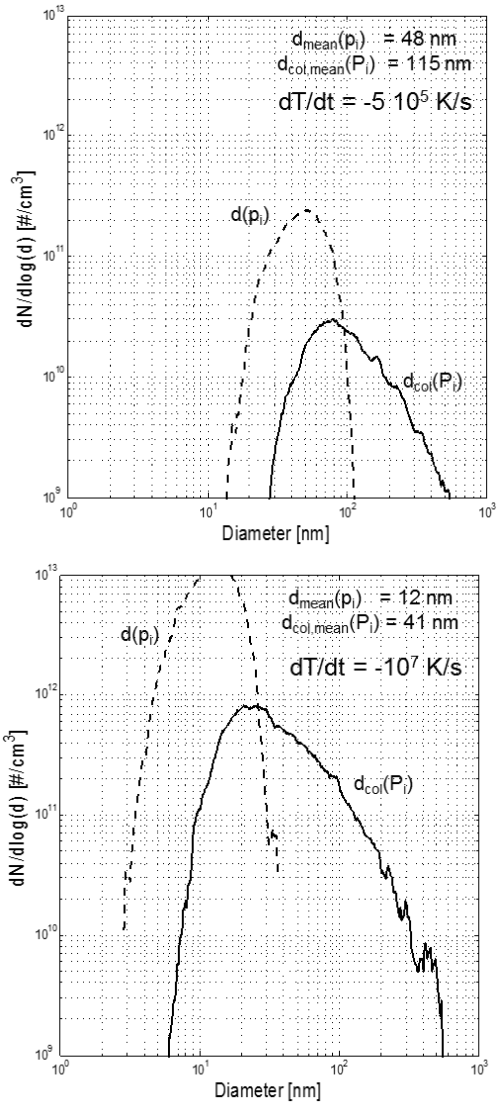

Figure 3 – Particle size distribution for two different quenching rates.

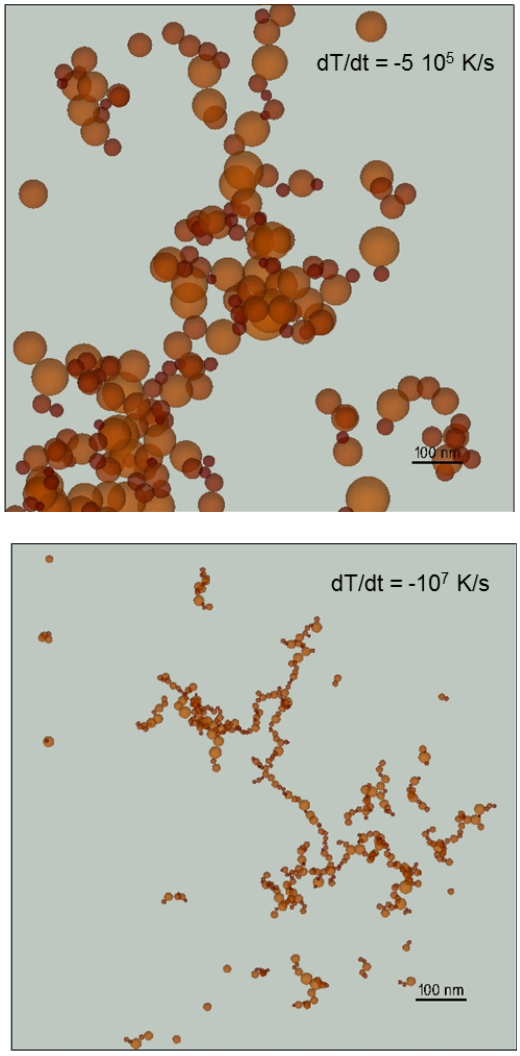

Figure 4 - TEM-style picture representing nanoparticles morphology at the end of the fume formation process (at T = 300 K) with primary particles coloured according to their composition.

The evolution of particulate phase for the case of -10^6 K/s cooling rate, together with the composition evolution, is represented in Fig. 5, showing a sequence of particulate samples as function of temperature, as predicted by the model. It's important to notice that coalescence upon coagulation of primary particles is the dominant phenomena up to 1800 K. At 1300 K it is possible to identify aggregates composed of more than two primary particles, because of a sintering time that is now reduced to values comparable with the collisional time.

4. Conclusions

Results show that both oxidation rate and nanoparticles size are affected by the cooling rate. In particular, the correlation between oxidation and cooling shows a non-linear behaviour. The reduction of particles size at higher quench rates predicted by this model is a well-known phenomenon, in agreement with modelling and experimental results in literature.

5. References

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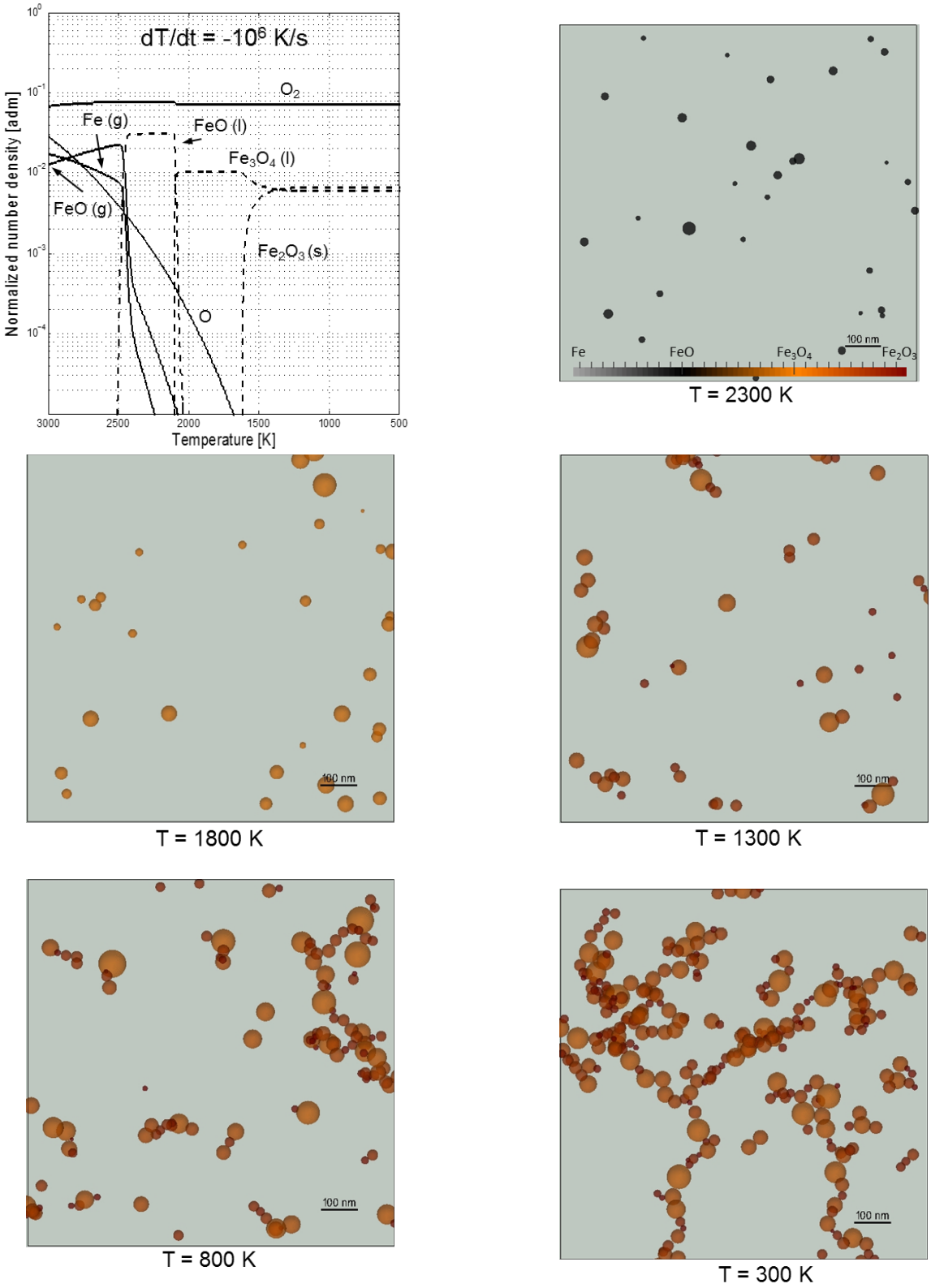

Figure 5 - TEM-style picture representing nanoparticles morphology during the quenching process.