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# Bimodal size distribution of primary particles in the plasma of welding fume: Coalescence of nuclei



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## ABSTRACT

Nucleation and growth of the nuclei in the thermal plasma of the ionized metal vapors of welding fume are studied. The coalescence of liquid droplets formed by the vapors' nucleation is calculated. There is a high flow rate of condensable atoms from the gas phase into the condensed phase caused by the nucleation and association of nuclei with the aggregated droplets. The fast association of the nuclei with aggregated droplets leads to a decrease in the condensable vapor supersaturation and to the termination of nucleation and start of the bimodal coalescence. As a result, in the beginning of phase transition, there is bimodal size distribution of the primary particles: the small-size primary particles formed by association between the nuclei (intramodal coalescence only), and the primary large-size particles formed by association between the aggregated droplets and the nuclei (intramodal and intermodal coalescence).

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## 1. Introduction

A low-temperature thermal dusty plasma is an aerosol at atmospheric or higher pressure and at temperatures of 1000–3000 K, which contains easily ionizable atoms of alkali metals as a natural impurity or in the form of specific additional agents. Ionization in a thermal plasma occurs due to collisions between electrons and alkali metal atoms, which are the basic suppliers of free electrons and singly charged positive ions. Therefore, a thermal plasma is strongly collisional unlike a low pressure gas–discharge plasma (Fortov et al., 2004). Examples of thermal collisional plasmas are the combustion plasma, which is formed in flames (Doroshenko et al., 2009; Poletaev & Florko, 2008), and the condensation zone of arc welding fume (Kobayashi et al., 1983). Both kinds of plasma represent ionized gas containing the vapor of metals and/or metal oxides, which are ready to condensate; therefore, as a result of volume condensation, such a plasma contains the dust as liquid droplets and, after the cooling and phase transition, as solid particles.

Such a plasma can be used in the technology of synthesis of nano-sized oxide particles with the required properties (Gonzalez et al., 2008; Kumfer et al., 2010; Seo & Hong, 2012). Thus, a new technology can be developed to obtain new materials. Flexible ceramics is an example of such new materials that can be used to make crucibles for melting metals, gas turbines, liners for jet and rocket motor tubes, resistance furnaces and ultra-high frequency furnaces (Laurvick & Singaraju, 2002). Another application of nano-sized particles is the production of fuel cells. Solid oxide fuel cells differ from other fuel cell technologies, because they are composed of all-solid-state materials, and as a result can operate at temperatures significantly higher than any other category of fuel cells (Singhal & Kendall, 2003).

The formation of condensed particles in the plasma occurs as a result of nucleation and growth of the nuclei (Girshick et al., 1993; LaViolette et al., 1996; Reist, 1984), and this process is strongly dependent on the plasma properties (Vishnyakov, 2008;

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Vishnyakov et al., 2011). The temperature in the condensation zone is higher than melting point of the condensable matter, therefore the nuclei are in the liquid state. The condensable vapor mixes with air and this vapor–gas mixture cools down, which leads to the beginning of growth of the nuclei. The growth of the nuclei occurs until the temperature reaches the melting point. The phase transition terminates the formation of the primary particles; secondary particles are formed as a result of further coagulation.

The dusty plasma of arc welding fume was investigated by Fuglsang et al. (2011), Asbach et al. (2009) and Brand et al. (2013). It was discovered that the primary particles are described by the bimodal size distribution. The paper by Fuglsang et al. (2011) demonstrated that particles of welding fume, collected by an Electrical Low Pressure Impactor, have a size of the first mode below 10 nm and a size of the second one is about 100 nm. But it should be noted that these particles are not the primary particles. Similar studies of the secondary particles (agglomerates of the primary particles) by Asbach et al. (2009) also demonstrated the bimodal size distribution with sizes of about 10 nm and 100–150 nm. The paper by Brand et al. (2013) presented the number-based size distributions of the particles of welding fume, where three modes can be observed: the particles of the first mode smaller than 6 nm, a size of the second mode is 20–30 nm and a size of the third mode is 100–200 nm. The investigation by Ennan et al. (2013) found two close modes of the secondary particles with sizes of  $\sim 200$  and  $\sim 300$  nm, and shown that these secondary particles can be a result of coagulation of the primary particles with sizes of  $\sim 2$  and  $\sim 20$  nm. The origin of the first mode is clear, it is the primary particles formed as a result of nuclei' growth. As to the second mode, one may assume that these particles are the result of droplets' coalescence (Tashiro et al., 2010).

This paper deals with the nucleation and growth of the nuclei occurring simultaneously with their coalescence; besides, the exchange processes between the particles and the plasma, where the nucleation occurs, are taken into account. The proposed calculation method allows defining the primary particle size distribution function in any moment of time.

## 2. Nucleation and growth of nuclei without coalescence

The calculation of heterogeneous ion-induced nucleation in the plasma by taking into account the interphase interaction, when nucleation depends on the electron exchange between the nucleus and the plasma, was done using the procedure suggested by Vishnyakov et al. (2013). The subject of modeling is the dusty plasma formed during cooling of the iron vapor stream from the temperature of 3000 K down to the melting point of iron (1800 K) as a result of mixing with the air. The vapor–gas mixture contains iron atoms and alkali metals as ionizable atoms in the following ratio:  $g_{Fe} = 0.36$ ,  $g_K = 0.06$ ,  $g_{Na} = 0.03$ , where  $g$  is the part by weight (the system under consideration corresponds to the plasma of shielded metal arc welding fume, Oprya et al., 2012).

The presence of the alkali metal atoms provides the content of ions with the equilibrium number density from  $3.6 \times 10^{15} \text{ cm}^{-3}$  (atom density is  $1.8 \times 10^{18} \text{ cm}^{-3}$ ), at the temperature of 3000 K, to  $7.5 \times 10^{12} \text{ cm}^{-3}$  (atom density is  $1.2 \times 10^{18} \text{ cm}^{-3}$ ), at the temperature of 1800 K. But real ion number density is less, because ions are the nucleation centers; for example at the temperature of 1800 K their number density is  $5 \times 10^{12} \text{ cm}^{-3}$  (atom density is  $6.5 \times 10^{17} \text{ cm}^{-3}$ , i.e. about  $5.5 \times 10^{17} \text{ cm}^{-3}$  atoms of alkali metals via ionization became a nucleus center). In the used procedure the ionization of alkali metal atoms due to the collisions and UV-radiation from the arc is considered. The electrons appear in the system not only due to the atoms' ionization, the thermionic emission and photoemission from the particles are also considered in the procedure. Therefore, the number of electron can be more than a number of ions, if the particles did not accumulate the major part of electrons. For example, at the temperature of 1800 K, the electron number density is  $5.4 \times 10^{12} \text{ cm}^{-3}$ .

As the vapor stream mixes with the air, the partial pressures of condensable vapors ( $P_i$ ) in the vapor–gas mixture and their supersaturations ( $S_i = P_i/P_{i,sat}$ ) can be calculated using the temperature and the vapor composition:

$$P_i = \frac{\delta m_i / \mu_i}{\delta m_0 / \mu_0 + \delta m_{air} / \mu_{air}} P, \quad (1)$$

where  $\delta m_i = g_i \delta m_0$  is the flow rate of the  $i$ th component;  $\delta m_0 = 10^{-2} \text{ g/s}$  is the mass flow rate of the vapors stream;  $\mu_i$  is the atomic mass of the  $i$ th component;  $\mu_0 = 26 \text{ g/mol}$  is the effective atomic mass of the vapors stream,  $\mu_{air}$  is the air molecular mass;  $P = 1.01 \times 10^5 \text{ Pa}$  is the atmospheric pressure;  $\delta m_{air}$  is the mass rate of entrainment of the air by the vapor stream, which is defined by the current temperature of the vapor–gas mixture  $T$ , the temperature of the environment  $T_\infty \sim 300 \text{ K}$  and the initial temperature  $T_0 \sim 3000 \text{ K}$ :

$$\delta m_{air} = \frac{T_0 - T}{T - T_\infty} \delta m_0. \quad (2)$$

The current total number density of iron atoms in the vapor without account of the condensed phase is determined using (1) via the perfect-gas relation:

$$N_{a0} = \frac{P_{Fe}}{k_B T}, \quad (3)$$

where  $k_B$  is the Boltzmann constant. Adjusted to take into account that some iron atoms are in the condensed phase, the number density of the iron atoms remaining in the gas phase is

$$N_a = N_{a0} - N_{ac},$$

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