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# A model for the particle mass yield in the aerosol synthesis of ultrafine monometallic nanoparticles by spark ablation

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ABSTRACT

In this work we present a systematic study of metallic nanoparticle production by spark ablation. We show how to adjust key process parameters to tune the obtained particle size, concentration and mass for various metals and carbon, which we produced in its amorphous form according to Raman spectroscopy, as the present experimental setup offers the possibly for a fast and easy exchange of the electrodes. For nearly all tested materials a mean particle size of below 5 nm can be obtained if proper process conditions are chosen. We show that energy efficiency of the spark ablation process can be increased by higher circuit capacitance and smaller electrode diameters. Furthermore, we correlate the particle mass production during spark ablation with a modified Fourier number. This allows us to compare expected mass yields for different elements and indicates that heat dissipation and especially conduction away from the spark region plays an important role for the particle production.

## 1. Introduction

During the last decades metallic nanoparticles have experienced increasing scientific attention due to unique properties that arise from their small particle size such as high curvature and percentage of surface atoms. Nanoparticles show higher luminescence, strength and hardness as the respective bulk materials (Gutsch et al., 2002). For their many advantageous properties, they pose an attractive material for the application in many fields such as catalysis (Dhas, Raj, & Gedanken, 1998), sensory devices (Mädler et al., 2006), microelectronics (Tseng & Chen, 2006) and optics (Huang et al., 1997).

Manufacturing methods for metallic nanoparticles can be divided into two general approaches: liquid phase-based processes and gas-phase synthesis. The former methods suffer from impurities in the produced material due to the necessity of using surfactants for stabilization against coagulation. In contrast to that, gas-phase synthesis allows for the production of ultra-pure nanoparticles of nearly every metal. Gas phase processes can be divided into two sub-classes: The first one comprises methods where a precursor substance undergoes decomposition to form either – in case of reducing reaction conditions – metallic or oxidized nanoparticles. Examples for the respective methods include flame spray pyrolysis, which is especially suitable for the production of complex mixed oxides (Kunzmann, Stanzel, Peukert, Costa, & Guldi, 2016; Stark, Mädler, Maciejewski, Pratsinis, & Baiker, 2003) and high production rates (Mueller, Mädler, & Pratsinis, 2003). It can also produce metallic nanoparticles under certain process conditions (Grass & Stark, 2006). Synthesis in plasma and hot-wall reactors also belongs to this group. The latter one is regularly used for the synthesis of metallic (Nasibulin, Kauppinen, Brown, & Jokiniemi, 2001) and also semiconducting multicomponent particles (Mehringer et al., 2014).

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The second sub-class consists of inert gas condensation processes. In such processes a metallic target is evaporated to form a vapor which is then quenched by an inert gas. This leads to a high supersaturation of the metal in the gas phase and consequently to its homogeneous nucleation and formation of metallic nanoparticles. Among these processes are for example arc discharge evaporation methods that already proved to be suitable for the production of metallic nanoparticles in a broad range of sizes and with reasonable long-term stability as it was shown by Chazelas, Coudert, Jarrige, and Fauchais (2006) and Förster, Wolfrum, and Peukert (2012). Also, the capability of this method to produce mixed metal particles has been demonstrated, for example for Al-Mn (Lee, Li, Choi, & Dong, 2010) or Al-Mg compounds (K. Akbari, Derakhshan, & Mirzaee, 2015). Furthermore, the evaporation of metal from a resistively heated wire can be harnessed to produce metallic nanoparticles as demonstrated for gold by Boies, Lei, Calder, Shin, and Girshick (2011) and mixed metallic nickel- and iron-based wires by Khan et al. (2014).

Spark ablation also belongs to the inert gas condensation processes. It has been harnessed to produce nanoparticles (Schwyn, Garwin, & Schmidt-Ott, 1988) for quite a long time. Spark ablation features some unique properties that make it especially suitable for the production of ultrafine metallic nanoparticles: The setup can be kept very simple and compact as it works at ambient temperature, only safety measures with respect to the high voltage used for the spark discharge need to be considered. Furthermore, the method can be applied to nearly all conductive or semi-conducting materials (Tabrizi, Ullmann, Vons, Lafont, & Schmidt-Ott, 2009). As only thin metal rods or wires are needed to form the electrodes, the material costs can be kept low even for noble metals. Mixed metallic particles can be produced by using alloyed electrodes (Tabrizi, Xu, van der Pers, Lafont, & Schmidt-Ott, 2009) or two electrodes of different materials (Byeon, Park, & Hwang, 2008). Spark ablation in liquids instead of gases is another possibility leading to complex materials such as $Bi_{0.5}Sb_{1.5}Te_3$ with good thermoelectric properties (Nguyen et al., 2012). Although spark ablation generally produces low quantities of particles, concepts for upscaling exist (Feng, Biskos, & Schmidt-Ott, 2015; Hontañón et al., 2013; Pfeiffer, Feng, & Schmidt-Ott, 2014).

Whereas numerous studies were carried out regarding the particle production and demonstration of the applicability of spark-produced nanoparticles (Isaac et al., 2015; Isaac, Valenti, Schmidt-Ott, & Biskos, 2016; Valenti, Dolat, Biskos, Schmidt-Ott, & Smith, 2015), yet many mechanistic aspects about particle formation and remain unclear. This works presents a discussion of important process parameters and their influence on the produced material as well as an attempt to better understand the material dependency of particle production.

## 2. Material and methods

The spark generator used in this work (Fig. 1) adapts the design already used in Förster, Thajudeen, Funk, and Peukert (2016) which based on the work of Schwyn et al. (1988) and Tabrizi et al. (2009). The setup is housed within a KF40 six-way cross (VACOM GmbH, Großlöbichau) equipped with two viewports at the front and back flanges to observe the spark discharge. Carrier gas enters the system through the bottom flange and passes the spark electrodes guided by a two-piece glass-made flow guide. The piece downstream the spark has an inner diameter of 8 mm and an effective length of 45 mm. The now particle laden carrier gas leaves the system via the top flange through a stainless steel tube with an inner diameter of 4 mm and is quenched at a fixed ratio of 1:4 after 80 mm of tubing. Stainless steel tubing and a conductive polymer hose with an inner diameter of 4 mm and a length of approximately 800 mm transport the aerosol to the SMPS.

Argon (99.999%, AIR LIQUIDE Deutschland GmbH) was used as carrier gas to ensure inert conditions during discharge and particle formation whereas nitrogen (99.999%, AIR LIQUIDE Deutschland GmbH) was used for quenching. The flow rates were adjusted by rotameters equipped with needle valves. According to the supplier, 2 ppm of oxygen is present as impurity in both gases. This is sufficient to oxidize non-noble material produced via spark ablation.

Electrode rods of aluminum, hafnium, copper, molybdenum, niobium, silver, tungsten, tin, zinc, titanium and carbon were supplied from MaTecK GmbH and had a purity of at least 99.9% except for hafnium (99%). Prior to usage, the electrode tips were shaped hemispherical so that the discharge preferably occurs in a very small area in the center of the electrode tips. By default,

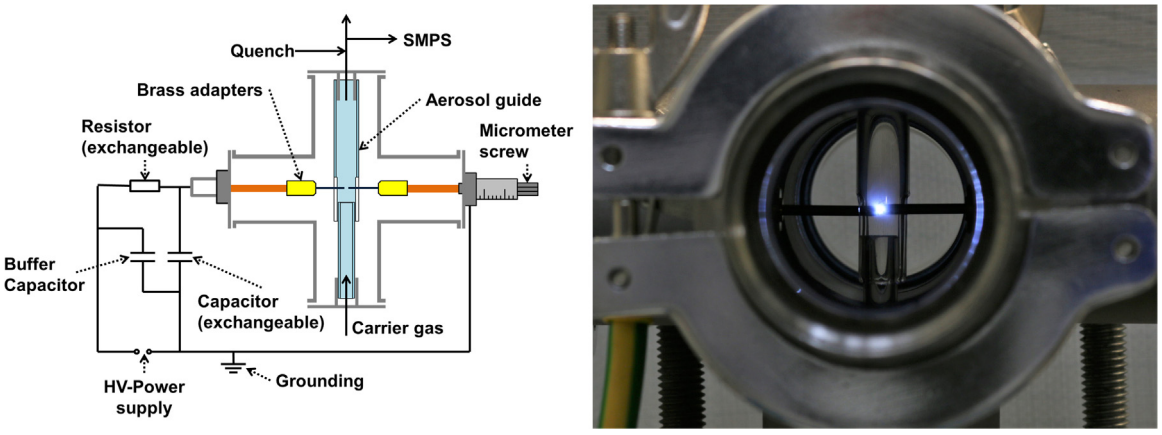


**Fig. 1.** Schematic drawing of the spark generator (left) photograph of a spark discharge at the setup (right).