Size Controlled Synthesis of Spherical Nanoparticles by Spark Discharge

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Spark discharge electrode ablation which was described firstly by Schwyn *et al* (1998) is a versatile method of nanoparticle production with a narrow size distribution. A spark generator basically consists of a pair of electrodes whereby repeated spark discharges take place over a gap within a mm range. Each spark generates a vapour cloud which condenses into nanoparticles. Meanwhile, an inert gas flow continuously flushes the gap for cooling and dilution. Spark generators are widely used in modern laboratories, and efforts are currently underway to scale up the process to achieve industrial production rates. Agglomerates are generated in most cases and thus lead to an agglomerate diameter, which are expected to overcome since the primary particle diameter is usually advantageous to control the properties of the product and individual sphere-like (primary) particles are required in various applications. Schwyn *et al* (1988) and Tabrizi *et al* (2009) have demonstrated that sufficient dilution can avoid agglomeration in the spark process.

In the present paper, we have systematically investigated the influence of process parameters on primary particle size. The ability to control the primary particle size of nanostructured materials is essential since it is a key variable in many thermal, mechanical and optical properties. Our present results are based on gold nanoparticles.

The inert gas dilutes and cools the vapour which forms the particles. In this work, parameters to be set are the repetition frequency *f*, the electrode gap distance *d*, the flow rate *Q* and the energy *E* provided per spark. The latter two parameters (*Q*, *E*) are expected to be of great importance as they directly influence the concentration and temperature in the vapour condensation process. A Scanning Mobility Particle Sizer system is applied to obtain the modal particle diameter. Different dilution rates lead to different sizes of spherical particle formation (Fig. 1). The inserts show electron micrographs with different parameters, indicating that particles smaller than ca. 5 nm have coalesced to form sphere-like shapes. According to the work of Koch *et al* (1990), it can be concluded that for particles larger than 5 nm $\tau_{coalescence} > \tau_{collision}$ is valid for the characteristic times for coalescence and collision. Spark energy also tunes the particle size shown in Figure 2. This is expected, since the energy of the spark directly influences the amount of material evaporated with relation to $\tau_{collision}$ as well as the temperature history, which controls $\tau_{coalescence}$. Under the conditions applied, the size of primary particles is reproducible and variable between 2.7 and 5 nm. Smaller particle sizes can be achieved, which can be decreased to the single atom (Peineke, 2009). Larger round particles can be produced by inducing coalescence of agglomerates by heating the aerosol stream (Kruis, 1998). The present progress has shown the potential to effectively control the size of primary particle in the spark generation process.

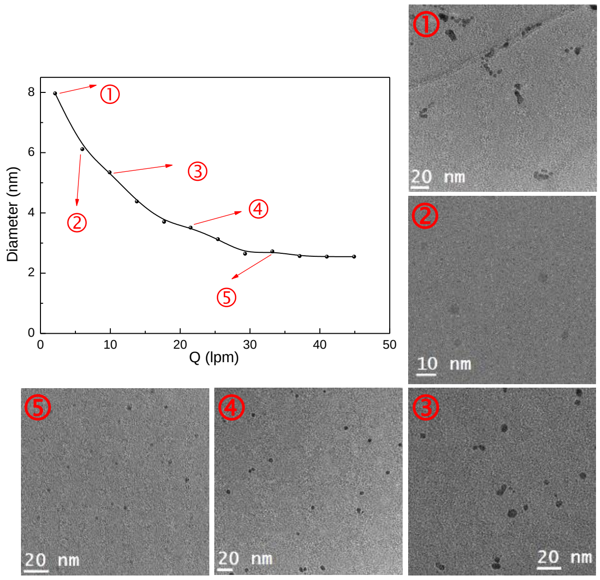

Figure 1. Particle diameter vs. flow rate (*E*=0.2623 mJ/spark, Spark duration=4 µs).

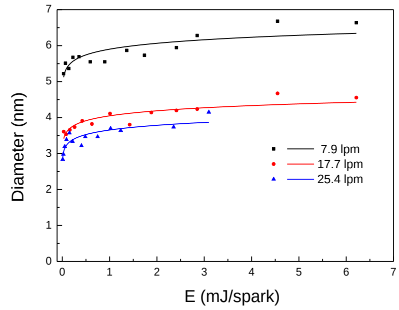

Figure 2. Particle diameters vs. *E*.

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