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The effect of circuit resistance on the particle output of a spark discharge nanoparticle generator

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ABSTRACT

The present study investigates how systematic variation of the resistance of the discharge loop influences the nanoparticle output and the electrode erosion of spark discharge generators (SDGs). The size distribution of the nanoparticles, as well as the mass loss of gold electrodes was recorded while varying the total resistance of the discharge loop. It was demonstrated that the characteristics of the aerosol nanoparticles produced by SDGs as well as the erosion rate of electrodes strongly depend on the total resistance even at small values. It was found that by increasing R_{total} from 0.7 Ω to 6 Ω, the modal diameter of the gold particles decreased from 39 nm to 16.5 nm with the concomitant decrease of the erosion rate. Our data allowed concluding that a fair part of the few ohms of total resistance typical to SDGs can easily originate from the electric circuit which affects the particle output. This also means that in addition to the usual control parameters, the total resistance should also be monitored in an SDG, conveniently via monitoring the current, for the purpose of maintaining stable and reproducible NP production. Moreover, circuit resistance can even be considered to be a practical control parameter, if certain size particles are to be produced by an SDG.

1. Introduction

Spark discharge nanoparticle generation is a technically simple, yet versatile and environmentally friendly technique for producing nanoparticles (NPs) of virtually any conducting material in the gas phase (Pfeiffer, Feng, & Schmidt-Ott, 2014; Schwyn, Garwin, & Schmidt-Ott, 1998). In the so-called spark discharge generators (SDGs), high-voltage and high-current, microsecond-long spark discharges are created between two electrodes in a controlled gas flow at atmospheric pressure. Due to the sparking, the electrode material is eroded and a vapor plume is formed between the electrodes, which is then converted to NPs via nucleation, condensation, coagulation, and aggregation (Borra, 2006; Feng, Biskos, & Schmidt-Ott, 2015). This process takes place in a quasi-continuous manner, i.e. sparking is maintained by charging a capacitor and discharging it via the spark gap in a repetitive manner.

The discharge loop of a typical SDG can considered to be an RLC circuit, the resistance and inductance of which typically originates from the cables, electrical connections, and the spark plasma itself (Meuller et al., 2012). The sparks are fed by the energy stored in the capacitor, which was shown to be one of the key parameters controlling particle formation in an SDG (Feng et al., 2016, Horvath and Gangl, 2003). Due to this primary role, the effect of the capacitance was studied in the literature and was shown to be correlated with the electrode erosion rate (Tabrizi et al., 2008). Since SDGs are usually built from conductive materials without

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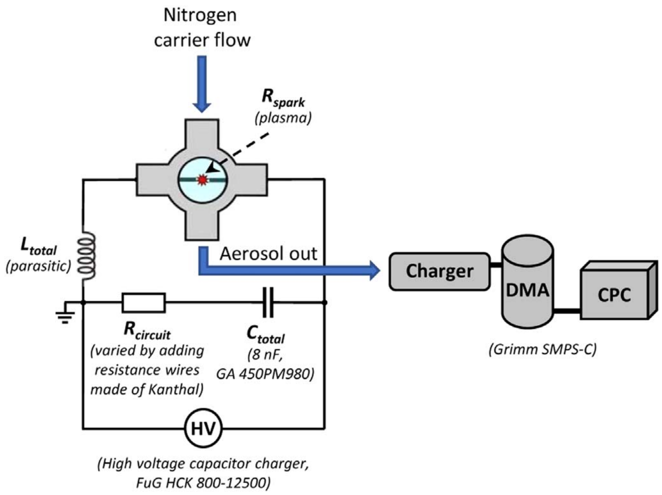

Fig. 1. Schematic view of the experimental setup.

intentionally adding resistors or inductors to the circuit, it is commonly assumed that – when highly conductive electrodes are used – the spark plasma is the main resistive component in the discharge loop (Vons et al., 2011). The resistance of the spark plasma cannot be measured directly in a common SDG, instead, the total resistance is usually calculated from the current or voltage waveform (Palomares, Kohut, Galbács, Engeln, & Geretovszky, 2016). The total resistance of SDGs used by different research groups, based on data available in the literature, is in the range of 1.5–5.0 Ω (Hontanon et al., 2013; Palomares et al., 2016; Tabrizi, Ullmann, Vons, Lafont, & Schmidt-Ott, 2009; Vons et al., 2011).

The present technical note reports about the observations we made in experiments in which we systematically varied the resistance of an SDG circuit and studied its effect on the characteristics of the NP output and the electrode erosion process. Our results provide a better understanding of the role and optimization possibilities of the physical parameters controlling the operation of SDGs.

2. Materials and methods

The SDG setup used in the experiments was formerly described in detail in a previous publication (Kohut, Galbács, Márton, & Geretovszky, 2017), hence here we only give a brief overview. The setup is schematically shown in Fig. 1. It is centered around a KF-sealed, DN-160 sized, cylindrical stainless steel chamber (Pfeiffer Vacuum GmbH) equipped with four, radially oriented KF-40 ports. A pair of cylindrical Au electrodes (99.9% purity, Kurt J. Lesker Co.) of 3.00 mm diameter was horizontally positioned and axially aligned inside the chamber. The inter-electrode distance (i.e. the gap size) was controlled by micropositioners (Model K150-BLM-1, MDC Vacuum Ltd.) and set to 1.0 mm for all results reported here. Nitrogen (99.995% purity, Messer Hungarogáz Ltd.) was employed as carrier gas, entering the chamber via a top KF-40 port (downward pointing “crossflow”). Experiments on the mass loss of electrodes were carried out in argon (99.996% purity, Messer Hungarogáz Ltd.) The gas flow rate was set to 1 slm by a mass flow controller (Model GFC16, Aalborg Inc.). All experiments were carried out at atmospheric pressure, maintained by a diaphragm pump and a needle valve, and monitored by a piezo-resistive pressure gauge (Model VD81, Pfeiffer Vacuum GmbH/Thyracont Vacuum Instruments GmbH). A monolithic, high voltage, pulse discharge capacitor (Model 450PM980, General Atomics) with a capacitance of 8 nF was connected to the spark gap and charged by a high voltage capacitor charging power supply (Model HCK 800–12500, FuG GmbH). The discharge of the capacitor between the electrodes commences when the voltage on the capacitor reaches the breakdown voltage in the electrode gap. The resulting spark discharge is a bipolar, oscillatory discharge. The repetition rate of the sparking (spark repetition rate, SRR) was kept constant at 100 Hz by controlling the charging current of the capacitor. The voltage and current waveforms in the discharge loop were recorded by a 200 MHz digital storage oscilloscope (Model DSOX2024A, Keysight Technologies Inc.) using a broad-band high voltage probe (Model P6015A, Tektronix, Inc.) and a current probe (Model 110, Pearson Electronics, Inc.), respectively.

The total resistance of the series RLC circuit (discharge loop) in the experiments was varied by inserting a piece of Kanthal resistance wire (FeCrAl alloy, Sandvik, Sweden) in between the capacitor and the spark gap, the length of which was varied between 15 and 90 cm. Our SDG initially had a resistance of ~0.7 Ω which was increased to 1.3, 2.1, 2.7 and 6.0 Ω in a stepwise fashion.

The size distribution of the generated Au NPs was measured downstream of the SDG by using a scanning mobility particle sizer (SMPS-C, Grimm Aerosol Technik GmbH) consisting of a condensation particle counter (Model No. 5.400) and a “Vienna-type” dynamic mobility analyzer (Model No. 5.500).

The erosion of the electrodes was measured gravimetrically by weighing the electrodes before and after two hours of sparking (equivalent to about 720,000 oscillating sparks delivered). The weighing of the electrodes was done on a semi-micro analytical balance (Model AB135-S/FACT, Mettler Toledo Kft.) that has an accuracy of 10 μg. Prior to use, the electrodes were thoroughly rinsed with ethanol and wiped clean with low-lint laboratory wipes (Kimtech Science, Kimberly Clark).