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# Surface features and energy considerations related to the erosion processes of Cu and Ni electrodes in a spark discharge nanoparticle generator

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## ABSTRACT

In the present study, the erosion of polished nickel and copper electrode surfaces (with an average roughness of ca. 4.5 nm) exposed to a low number $(1-3)$ of sparks in a spark discharge nanoparticle generator were investigated with the purpose of better understanding the utilization of the energy pumped into the electrodes as well as the processes leading to the generation of aerosol nanoparticles. It was shown that even a single oscillatory discharge creates complex morphological changes on the electrode surfaces. Three main erosion features were identified (referred to as *craters*, *undulated areas* and *dendritic areas)* and characterized by optical, confocal laser scanning and atomic force microscopy. Their potential formation mechanisms are also discussed. By estimating the total energy needed to melt the electrode material corresponding to the volume of the craters individually identified and associated with the first half-cycle of the discharge, it was shown that the electric energy pumped into the electrodes in the form of Joule heating is mostly consumed by the melting of the electrode material, which is followed by re-solidification. Hence, only a small fraction of the electrode material is actually evaporated into the ambient gas. This explains the very low value of the energy-efficiency in the commonly used Jones model.

Our results indicate that only a part of the material that leaves the electrode surface is actually converted into an aerosol. The input to the aerosol formation is either vapor or molten material ejected from craters. Ejected molten material can lead to the formation of micrometer-sized aerosol particles ("*splashing particles*"). Some of the metal vapor is deposited on the surface in the vicinity of craters forming dendritic areas, whereas only a fraction of the metal vapor is carried away by the ambient gas and may form aerosol nanoparticles. This study clearly indicates that erosion processes in spark discharge nanoparticle generators are very complex and can not be reasonably described by simplified erosion models.

## 1. Introduction

Nowadays there seems to be a great and continuously growing need in the industry for the versatile, reproducible and mass production of nanoparticles (NPs). One of the few techniques that can meet the requirements is the generation of NPs by means of

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electrical spark discharges (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, made of practically any conducting or semi-conducting material, in a controlled inert gas flow at atmospheric pressure in order to produce high-purity NPs in an environmentally friendly and up-scalable way (Meuller et al., 2012; Pfeiffer, Feng, & Schmidt-Ott, 2014). Although the fundamentals of the generation process are not yet fully understood, the overall process is that the spark plasma erodes the electrode material and produces vapor, which is converted to NPs via nucleation, condensation, coagulation and aggregation (Borra, 2006; Feng, Biskos, & Schmidt-Ott, 2015) facilitated by the carrier gas. Consequently, a key element of the process is electrode erosion.

The electrode erosion in an SDG is usually described by the simple model of vaporization of the electrode material in a small spot created by the spark plasma channel (Feng, Huang et al., 2016; Muntean, Wagner, Meyer, & Seipenbusch, 2016; Pfeiffer et al., 2014; Tabrizi, Ullmann, Vons, Lafont, & Schmidt-Ott, 2009; Feng, Ramlawi, Biskos, & Schmidt-ott, 2018). By assuming that the energy delivered by the spark channel is in balance with the energy output represented by thermal conduction and radiation, the mass evaporated from such a spot can be calculated (Jones, 1950); this is the so-called Jones model. The Jones model requires an experimentally determined energy-efficiency factor, which has a remarkably low value – in the order of 0.1% – in case of a traditional SDG (Feng, Huang et al., 2016). By incorporating the energy efficiency factor (as well as other experimentally determined parameters), the mass derived from the energy balance gives a reasonably good estimate of the material eroded in an SDG at varying inter-electrode distances (Feng et al., 2015) and for several electrode materials (Muntean et al., 2016). However, the highly simplified character of this approach is well reflected by the fact that it erroneously predicts the relative erosion of certain materials, e.g. gold and silver (Tabrizi, Ullmann et al., 2009) and cannot explain the origin of the very low value of the energy-efficiency factor.

The electrode erosion in the SDG is the source of atomic vapor which feeds NP formation. Therefore the erosion process is usually investigated indirectly through the examination of the generated particles (e.g. Horvath & Gangl, 2003; Feng, Wong & Hong, 2016). Direct measurements investigating the erosion process were performed exclusively on the multi-spark level – when the electrode surface is treated by thousands or even millions of sparks – usually by gravimetry (by measuring the mass loss of the electrodes due to the sparking (Tabrizi, Ullmann et al., 2009; Muntean et al., 2016)). The changes of the electrode surface morphology after prolonged sparking in an SDG was only recently examined by the present authors, and the formation of self-ordered patterns was reported on various electrode materials (Wagner, Kohut, Geretovszky, Seipenbusch, & Galbacs, 2016). Contrary to the limited experimental data on the electrode erosion (especially dealing with the morphological changes of the surface) in an SDG, extensive research was conducted on the electrode erosion in related areas such as cathodic arc discharges (Anders, 2009), microelectric discharge machining (Feng, Wong et al., 2016) or spectroanalytical spark discharges (Brewer & Walters, 1969; Ekimoff & Walters, 1981). A comprehensive overview on the formation of surface patterns in case of various types of discharges, such as DC glow, arc, gliding arc or barrier discharges is given in a recently published cluster issue (Benilov & Kogelschatz, 2014).

Literature results, including our recent results (Wagner et al., 2016), suggest that the appealingly simple picture of the Jones model, namely that electrode erosion in an SDG takes place at a single spot heated by the spark channel, is an oversimplified one. In order to gain a better understanding of the utilization of the energy pumped into the electrodes as well as the processes leading to the generation of aerosol nanoparticles, the erosion of the surface of the electrodes is investigated on a single-spark level in the present study. The erosion patterns on nickel and copper electrodes are characterized by optical, confocal laser scanning and atomic force microscopy and their potential formation mechanisms are also discussed.

## 2. Materials and methods

### 2.1. Experimental setup

The flowchart of the setup is schematically depicted in Fig. 1. The chamber geometry was optimized for versatility and NP production (www.buonapart-e.eu), and the chamber itself was manufactured by Pfeiffer Vacuum GmbH. It is a DN-160 sized, cylindrical stainless steel chamber equipped with four, radially oriented KF-40 ports. The chamber was set up in an upright position, in

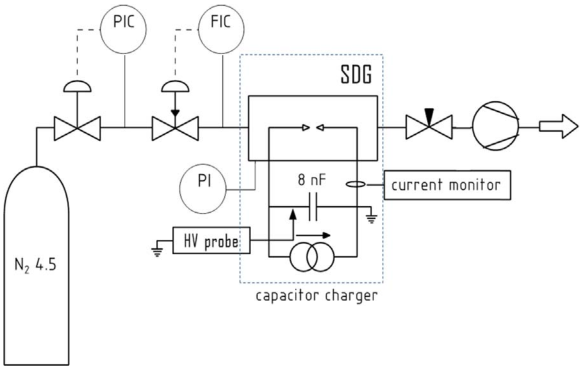

**Fig. 1.** Schematic picture of the experimental setup. The abbreviations PIC, FIC, PI, and HV probe stand for pressure indicator controller, flow indicator controller, pressure indicator, and high voltage probe, respectively.