Advanced Powder Technology xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Original Research Paper

Analysis of multi-scale Ni particles generated by ultrasonic aided electrical discharge erosion in pure water

Yifan Liu, Kunlun Zhu, Xianglong Li *, Faming Lin, Yan Li

College of Manufacturing Science and Engineering, Sichuan University, Chengdu 610065, China

A R T I C L E I N F O

Article history:
Received 11 July 2017
Received in revised form 18 December 2017
Accepted 10 January 2018
Available online xxxx

Keywords:
Ultrasonic aided electrical discharge erosion
Colloids
Multi-scale
Morphology and crystal structure
Size distribution

A B S T R A C T

Electrical discharge erosion is widely applied in the fabrication process of the metallic particles in liquids. The Ultrasonic aided electrical discharge erosion is based on the spark discharge in pure water. The synthesized colloids were classified in accordance with the nano size and micron size. The higher magnifications of morphology, chemical compositions, the crystal structure of the multi-scale particles were observed and analyzed by SEM, TEM, EDS, and XRD. It is verified that ultrasonic wave influenced the morphology of micro/nanoparticles and the roughness of inner and external surfaces of hollow microparticles. Besides, based on results of EDS, XRD, and Quantitative phase analysis, it is confirmed that nickel oxide was detected only on the surface of microparticles but the nickel oxide was easily obtained when nanoparticles were formed. In addition, ultrasound wave affected the oxidation reaction in both scales but the reaction was remarkably enhanced on nanoparticles. The DLS and LPSA were used to measure the size distributions for the nano and micron scale, respectively. The D-Values of both conditions shown that the ultrasound has an enhanced effect on decreasing the size distribution in both scales.

1. Introduction

In the past two decades, the requirement of micro/nano-scale metal particles has rapidly increased in many application areas such as semiconductors, solar cells, catalyst, 3-D printing (printing electronic devices and additive manufacturing) and powder metallurgy [1–4]. Generally, metal particles with the size of nano or micro size possess unique or improved physical, thermal, electrical and chemical properties compared with their corresponding bulk materials. Therefore, synthesis of multi-scale metal particles has drawn more attention in the past years [5,6]. Electrical discharge erosion is a physical method that has been developed to fabricate various metal particles in micro/nanoscale. Compared to the conventional chemical methods [7], electrical discharge erosion method is widely utilized due to its low-cost, eco-friendly, and time-saving properties. During the discharge erosion process, metallic particles are formed in different ambient cryogenic media like condensed gas (liquid nitrogen) [8], flowing gas [9] and liquid [10]. Generally, the synthesized metal particles have a wide particle size distribution ranging from a few tens of nanometers to several tens of micrometers. The multi-scale (nano- and micro- scale) distribution of synthesized particles is highly influenced by the dielectric mediums, energy input and discharge time. Especially, the wide particle size distribution is easy to be observed when liquid serves as the working medium. Some researchers applied electrical discharge erosion method to produce metallic particles in different solutions. Dvornik [11] used spark erosion to fabricate nanostructured WC-Co particles in distilled water. M.R. Shabgard [12] synthesized nano-structured tungsten carbide (WC) powder using electro-discharge process with tungsten and graphite electrodes, which were submerged in two different dielectrics (kerosene and deionized water). R.K. Sahu et al. [13] employed micro-electrical discharge to prepare the copper micro/nanoparticles under in de-ionized water mixed the stabilizers like polyvinyl alcohol (PVA) and polyethylene glycol (PEG). Tseng et al. [14–16] fabricated the silver gold nanoparticles in different dielectric solutions using the spark erosion. Their devices for generation particles in liquids are based on the electrical discharge machining (EDM). In this process of EDM, low voltages (less than 200 V) and large currents (magnitude of tens Ampere) are applied to supply the energy and trigger the discharge process. M. Mardanian [17] used electrical discharge with high voltage (12 KV) to treat a mixture of copper, indium, and selenium powders to synthesize CIS nanoparticles in pure ethanol. In addition, ultrasonic technology was usually to assisting synthesis of particles. Chang [18] used arc

---

* Corresponding author.
E-mail address: lixianglong@scu.edu.cn (X. Li).

https://doi.org/10.1016/j.apt.2018.01.003
0921-8831/© 2018 The Society of Powder Technology Japan. Published by Elsevier B.V. and The Society of Powder Technology Japan. All rights reserved.

Please cite this article in press as: Y. Liu et al., Analysis of multi-scale Ni particles generated by ultrasonic aided electrical discharge erosion in pure water, Advanced Powder Technology (2018), https://doi.org/10.1016/j.apt.2018.01.003

discharge assisted-ultrasonic vibration to obtain TiO_2 nanoparticles in deionized water. Ghomi [19] synthesized gold nanoparticles in deionized water and pure ethanol using ultrasonic-assisted spark discharge and revealed that the ultrasonic wave increased the shape uniformity of the nanoparticles and decreased their size. Liu et al. [20,21] also used the ultrasound-assisted spark discharge erosion to fabricate Ni micro-particles. We mainly focused on generating Ni micro-particles in different liquids and investigated the influence of different electrical parameters and liquids on the particle size distribution as well as purity of particles. Apart from these, the formation mechanism of different particle structures (mainly in the microscale) and rate of hollow structural Ni particles also were studied in these works. Although we found that the ultrasound provided a positive effect on narrowing the size distribution of micro-particles and improving the proportion of hollow particles, the drawback of these works [20,21] is that nanoparticles were neglected which suspended in the colloidal solution and were difficult to collect at the end of the process by using the magnet.

In this paper, we chose a new collection method which was described in Section 2. We pay more attention to the formation mechanism of different scale particles and explain the formation process through a high magnification observation like SEM (scanning electron microscopy) and TEM (transmission electron microscopy). The crystal structures of multi-scale particles were identified by XRD (X-ray diffraction) and SAED (selected area electron diffraction). The chemical compositions were detected by EDS (energy dispersive spectroscopy). The size distributions of multi-scale particles were measured by Dynamic Light Scattering (DLS) for nanoscale and Laser Particle Size Analyzer (LPSA) for micron-scale.

2. Experimental theory and methods

The electric discharge generator is based on a controlled electrical discharge between two electrodes. The electrical circuit used on the electric discharge generator is equivalent to a simple RC circuit (Fig. 1a), where a capacitor placed in parallel to the gap between two sparking electrodes and charged by a constant current source with a low voltage (less than 200 V). When the electrode gap reaches a particular, very small size, the applied voltage on the capacitor exceeds the breakdown voltage of the dielectric solutions. Then, the capacitor is discharged over the electrode gap and the dielectric molecule is ionized by the electric field, which results in an increase of the concentration of electrons and ions in the dielectric solutions between the spark gap. Subsequently, the conductive channel (plasma, coloring in red in Fig. 1b and c) is formed due to the high concentration of matters. The thermal energy enables local to superheat on the surface of electrodes (coloring in orange in Fig. 1b and c), and the temperature rises locally to tens of thousands of degrees' Kelvin leading to heating, melting, boiling and evaporation of electrode material [22,23]. The dielectric liquid between the gap is simultaneously evaporated and molecules are dissociated into hydrogen and oxygen, resulting in a bubble (coloring in light blue in Fig. 1b and c) filled with gas and vaporized dielectric medium [24]. The molten drops and vaporized clusters are rapidly quenched and sintered to form particles. When the plasma collapses at the end of the discharge, the superheated region boils violently and unstably, leading to ejecting metallic clusters and molten droplets (coloring in orange in Fig. 1b and c). The ejections pass straightly through the gas bubble and penetrate the bubble wall (sheath) into the dielectric liquid (coloring in blue in Fig. 1b and c). The gap phenomena between the anode and the cathode regions can be simplified and described in Fig. 1(b) and (c).

It is noted that the inner pressure of the bubble is extremely high (280 Mpa) and the boundary between bubble and liquid expands with a velocity of several tens m/s [25,26]. The diameter of the bubble reaches several millimeters [23]. Obviously, there are opportunities to form different structures (spherical particles, non-spherical fractals, hollow particles, solid particles) as well as different scales due to the different phases of erosions and surrounding contents around the spark discharge zone. The different forming routes were described in our previous publication [21] through a hypothesis, i.e. the formation mechanisms were simply hypothesized by observing of visible morphology and structure of Ni particles. Actually, the difference in the morphology and scale of particles indicates the difference in the mechanisms of particles formation. Here, the different structures are interpreted on micro and nanoscale. Fig. 2 shows the schematic diagram of ultrasound-aided electric discharge erosion experimental process built to produce and analyze Ni multi-scale particles. The system consists of a power supply, a servo system, a pure water supply, ultrasound generator, a sample collector and a post-analyzing sys-

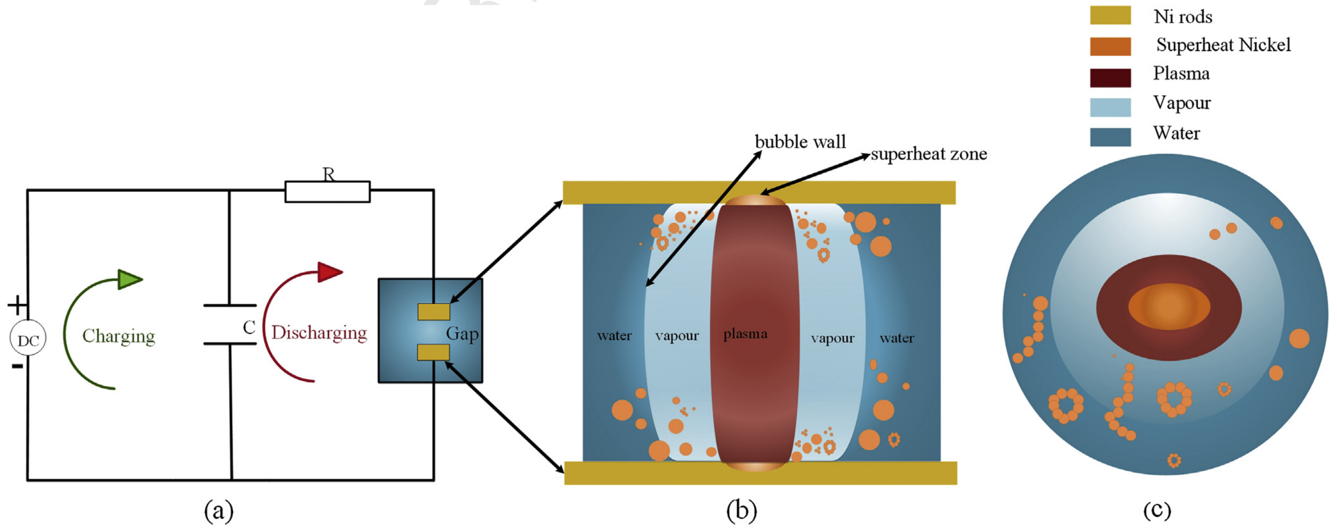

Fig. 1. Schematic representation of electric discharge erosion: (a) The spark as an RC circuit; (b) Schematic view of discharge gap (orange: Nickel; yellow: Nickel rods; dark red: plasma; light blue: vapor bubble; blue: dielectric solution); (c) Vertical view of discharge gap. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Please cite this article in press as: Y. Liu et al., Analysis of multi-scale Ni particles generated by ultrasonic aided electrical discharge erosion in pure water, Advanced Powder Technology (2018), https://doi.org/10.1016/j.apt.2018.01.003