Electrokinetic Stability and Size Modeling of Discharge Product Particles

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This study characterizes the colloidal stability and aggregation behavior of carbon discharge product particles synthesized via arc discharge. The Zeta potential distribution was successfully modeled as a mixture of two normal distributions with mean values of 50.02 mV and 18.74 mV. These results indicate that although the suspension is electrostatically stabilized by a predominant net positive charge, it possesses significant charge heterogeneity. The particle size distribution was modeled using the Fréchet distribution, identifying a baseline mode of approximately 2.7 um in the non-sonicated state. Experimental analysis revealed that sonication triggers stress-induced aggregation rather than dispersion, increasing the mode to 4.0 um and the mean particle size toward 10 um due to the disruption of the stabilizing electrical double layer. These findings suggest that the material is dimensionally suitable for infiltrating fibrous substrates in composites, but the fabrication process must employ low-energy techniques to prevent undesirable agglomeration and maintain optimal dispersion.

Keywords: carbon discharge product, particle, zeta potential, infiltration, size distribution, colloidal stability

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I. INTRODUCTION

The Zeta potential [1] is the main characteristic of colloidal system defined as the potential difference between the fluid layer adsorbed on solid particulates (hereafter referred to as particles) and the bulk phase of the medium. This potential determines the surface electrostatic charge on suspended particles. The surface charge of particles is one of important characteristics determining the stability of dispersed system as well as their surface chemical activity which is thought to be the important characteristics for the filtration of particles in fabric fibers. Also, the Zeta potential becomes the key factor to assess the behavior and reactivity of nano particles [2, 3]. In first, it should be noted that the Zeta potential is the characteristics determining the solution or aqueous medium containing the particles rather than the characteristics of particles itself. The potential can have positive and negative values indicating the particles in liquid environment (suspension) possess a charge of positive and negative type. Other than that, considering the values of Zeta potential, there is no significant difference between positive and negative Zeta potential. But, depending on type of aqueous medium especially on pH of medium, the potential can change its sign. Although the pH of the prepared carbon suspension was not directly measured in this work, carbon particles synthesized by arc discharge typically exhibit a near-neutral to mildly acidic surface chemistry in aqueous media due to the formation of oxygen-containing functional groups (e.g.,–COOH, –OH, C=O), resulting in a suspension pH commonly reported in the range of 5.5–6.8. Carbon formed in aqueous arc discharge interacts with water during synthesis, resulting in oxygenated surfaces associated with weakly acidic pH conditions [3].

Beyond assessing colloidal stability and reactivity of nano particles, Zeta-potential analysis has recently been shown to play a key role in guiding the design and development of functional materials, particularly in catalysis and surface-engineering applications [4].

Zeta potential can be determined using a range of measurement approaches, which are generally classified into three principal categories: electrokinetic mobility methods, electroacoustic methods, and streaming potential or streaming current methods, each encompassing multiple specific measurement techniques. In this work, Zeta potential was measured using a ZEECOM ZC-3000 analyzer (Microtec Co., Ltd., Chiba, Japan) at Laboratory of New materials, NUM. The instrument utilizes an electrophoretic mobility method, whereby a known electric field is applied across a suspension and the velocity of particle migration is measured via image-based tracking [5]. The resultant electrophoretic mobility is converted to Zeta potential using the Smoluchowski approximation, under the assumption of a thin electrical double layer [6, 7]. The measurement cell comprises quartz walls and palladium electrodes with defined spacing, ensuring controlled electric

field conditions and reproducible mobility determination. So, the measured value of the Zeta potential, ζ is calculated by following formula embedded [5] in the device software

$$\zeta = 9 \cdot 10^7 \cdot \frac{4\pi\eta}{\varepsilon} U\ [mV]$$

where: η-viscosity of solution, ε-dielectric constant, and U is electrophoretic mobility which is expressed as follows

$$U = \frac{v}{V/L}$$

(where v-speed of particle, V-applied voltage in Volts (V) , L-distance of electrodes).

When the carbon containing particles are used as microwave absorbing object in covering material, it should be infiltrated in the base material like fabrics. The strength of bonding of particles to the fabric fibers when the absorbing additional materials are infiltrated in fabrics depends on this value. The overall goal of this study is to establish a clear correlation between the material's surface chemistry and its macroscopic aggregation tendency, thereby providing fundamental data necessary for optimizing the composite infiltration process.

All data processing, statistical fitting (e.g., mixture models and Fréchet distribution), and visualization of the probability density distributions presented in this study were performed using the Wolfram Language programming environment within Mathematica, Version 12 [8].

II. SAMPLE PREPARATION, MEASUREMENT AND DATA PROCESSING

We have measured the Zeta potential of a sample which was produced by graphite arc discharge [9]. This product exists in a strong solid form of layered structures [10]. The sample was prepared from graphite used in arc discharge as electrodes. Initially, the solid material was mechanically ground using a porcelain mortar and pestle to reduce particle size. The ground material was then dispersed in ethanol and allowed to soak for 1–3 hours, followed by a second round of grinding. To remove residual ethanol, the sample was washed three times with deionized water and centrifuged at approximately 5000 rpm for 10 minutes, collecting the supernatant containing the finer fraction.

For suspension preparation, the recovered particle fraction was re-dispersed twice in deionized water, and serial dilutions were performed to obtain target concentrations suitable for analysis. To promote particle dispersion, the suspension was sonicated in an ultrasonic bath (35–40 kHz, 20 min), where acoustic cavitation generates localized micro-jets and shear forces, aiding in the breakup of weakly bound particle clusters and improves colloidal uniformity of aqueous sample. When necessary, additional probe sonication (1–2 min, pulsed mode) was applied to ensure further deagglomeration. Final suspension concentrations were adjusted to 100–500 mg/l for measurements.

Zeta potential measurements of the prepared samples were carried out using a ZEECOM ZC-3000 analyzer [5] at a controlled temperature of 22 ± 1 °C. Each sample was measured multiple times in order to obtain statistically reliable average values. The instrument simultaneously provides measurement results of several quantities for individual particles, including zeta potential, electrophoretic mobility, particle size, and associated statistical quantities, which are recorded in raw data sheet.

During the measurement, particle displacement and velocity under an applied electric field were directly measured by optical tracking. The electrophoretic mobility was calculated from the measured particle velocity, and the corresponding zeta potential values were subsequently determined by the instrument software. Statistical parameters, such as the standard deviation and coefficient of variation, were obtained from multiple particle trajectories.

The measured quantities were extracted from the raw data and subjected to further statistical analysis in which distribution of quantities, expressed by probability density function (PDF) is mainly identified. Distributions of the relevant quantities were analyzed by constructing histograms (with probability density) and determining the corresponding PDF through fitting procedures with maximum likelihood estimation on Mathematica [11].

III. RESULTS AND DISCUSSION

The histogram and probability density distribution of the Zeta potential values for the particles in aqueous sample was shown in Fig. 1. The distribution exhibits a mixture of two normal distributions, for which the PDF is given in common literatures as follows

$$\mathcal{N}(\mu,\sigma) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{(x-\mu)^2}{2\sigma^2}\right),$$

where: μ-mean value, σ-standard deviation. These distributions possess weights 0.16 and 0.84 in the mixed distribution and corresponding parameters are shown in format $\mathcal{N}(\mu,\sigma)$ in figure's legends (Fig. 1).

One of them has the mean value 18.74 mV with the standard deviation of 25.34 mV, whereas the other has 50.02 mV with the 9.64 mV standard deviation. These results tell us that the majority of particles possess positive potentials. The presence of two

distinct distributions indicates that the sample contains at least two particle subpopulations with different surface charge states. The existence of negatively charged particles alongside the positive ones leads to hetero-agglomeration, where oppositely charged particles attract each other to form larger clusters.

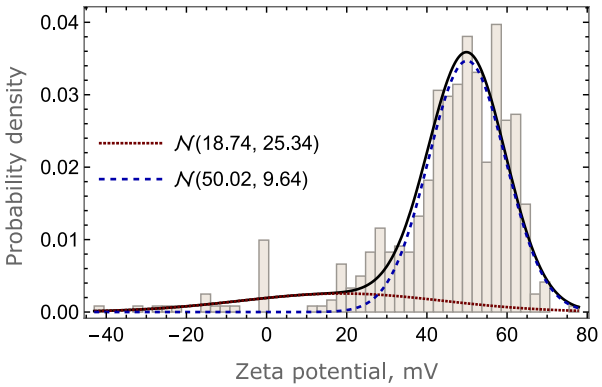

Fig. 1 Histogram (light brown bars) and probability density distribution of Zeta potential (solid black line) for the particles in suspension (mixtures are displayed by dotted and dashed lines)

However, the net excess of positively charged particles provides sufficient electrostatic repulsion to maintain the remaining material in a stable, dispersed state. Correlation analysis reveals no significant relationship between particle size and zeta potential. However, smaller particles exhibit a tendency toward higher surface charge magnitudes, which likely contributes to their enhanced colloidal stability through increased electrostatic repulsion. We should notice that the surface charge of particle may be calculated from their Zeta potential. Because Zeta potential is directly related to surface charge density, the electrokinetic data also provides indirect information on the surface chemistry of the particles.

Also, we can perform statistical analysis on size data of particles in the same way to previous analysis. The histogram and probability density distribution of particle sizes in the sample was shown in Fig. 2 for short duration (2-3 mins) of sonication. This statistical analysis reveals that size distribution of particles in aqueous medium is generally governed by the Fréchet distribution [12], PDF of which is given in common reliable literatures e.g. [12-13] as follows:

$$f(\alpha,\beta,\mu) = (x-\mu)^{-\alpha-1}\mathrm{e}^{-((x-\mu)/\beta)^{-\alpha}}$$

where: α-shape parameter, β-scale parameter, and μ-location parameter.

We obtain the size distribution given as $f[3.38, 3.70, -0.28]$. The shape parameter α is the most critical parameter in the Fréchet distribution as it controls the heaviness of the right-hand tail of the distribution. A value of $\alpha = 3.38$ indicates a moderately heavy-tailed distribution. Since the Fréchet distribution is often used to model extreme values (such as the largest particle size observed), this value suggests that the probability of observing very large particles is non-negligible, but the tail is not excessively heavy (which would correspond to $\alpha < 2$).

The scale parameter β determines the distribution's overall size and dispersion along the horizontal axis that represents the particle size variable. $\beta = 3.70$ um acts as a characteristic unit of size for the distribution. When combined with the shape and location parameters, this value helps center and spread the distribution to match the measured data. The Fréchet distribution uses the location parameter μ, to define the distribution's theoretical lower bound. The fitting value of this parameter was $\mu = -0.28\ um$ for this distribution.

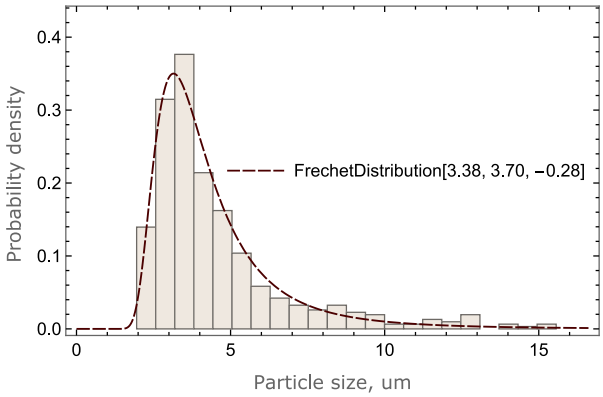

Fig.2 Histogram and probability density distribution of particle size in aqueous medium

Since particle size must be positive, the negative value of μ is a mathematical artifact of the fitting process. It indicates that the smallest particles measured are close to the detection limit near zero and the model uses this slight negative shift to optimize the fit across the entire range of measured values. In fact, the observed minimum value of particle size in suspension is $\mu = 1.95\ \mu m$. Also, the majority of particles are almost unimodal size ranging in 2-5 um in size. These are is consistent with the measurements of particle size on NANOPHOX.

Effect of Sonication on Particle Size

The stability was also quantitatively assessed by tracking changes in the size distribution upon sonication. We considered the particle size distributions of suspension in three cases (i) non-sonicated, (ii) short sonication, (iii) long sonication for 20-25 mins (Fig. 3). Three cases were analyzed using the Fréchet parameters.

In non-sonicated case (1-purple dotted line in Fig. 3), it represents the best dispersion, defined by a lower $\beta = 1.78$ and a tightly centered mode (2.7

um). For short sonication (2-darker red dashed line in Fig. 3), one can conclude that mechanical stress led to a rapid initial aggregation, and it is evidenced by the increase in the mode (3.2 um) and a large increase in spread ($\beta = 3.70$). This phase also correlates with the experimental observation that the mean size significantly increased up to several um after sonication. Long Sonication (3-darker green dot dashed line in Fig. 3) suggests that extended energy input promotes further aggregation, resulting in the broadest distribution ($\beta = 3.58$) and the largest mode (4.0 um). Although the scale parameter for long sonication is slightly lower than the short sonication case (3.58 and 3.70), the shape (α) and location (μ) parameters drives the distribution further to the right, resulting in the maximum observed mode of 4.0 um. This confirms that extended energy input promotes larger, more stable aggregate formation.

This quantitative analysis confirms that the primary mechanism during sonication is stress-induced aggregation. The mechanical energy momentarily disrupts the stabilizing electrical double layer, allowing the strong attractive forces between the oppositely charged species (as revealed by the Zeta potential analysis) to dominate, forming persistent aggregates.

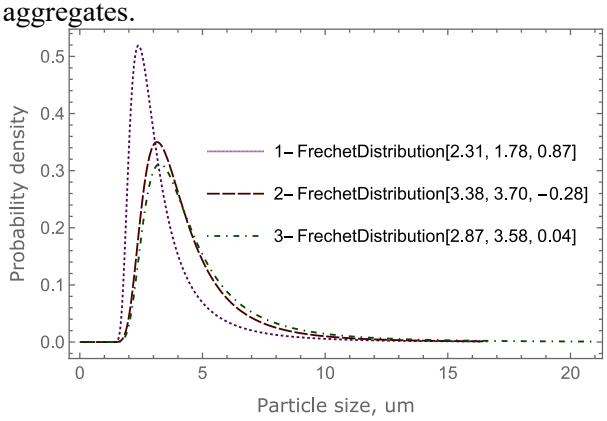

Fig. 3 Particle size distributions for non-sonicated (1-dotted purple), short sonication (2-dashed red) and long sonication (3-dot-dashed green) cases

Even though no statistically significant overall linear correlation was found between particle size and Zeta potential, the observed trend suggests that smaller particles exhibit a tendency toward higher absolute surface charge. These are the most electrostatically active components and are the primary drivers for the aggregation mechanism confirmed during sonication.

The fine scale and high dispersion potential (mode~2.7 um) are vital. Considering the material's intended use in fibrous composites for electromagnetic wave absorption, this dimensional compatibility is deemed highly appropriate for uniform integration and maximizing functional performance.

4. CONCLUSION

In conclusion, this research establishes that the carbon discharge products exhibit colloidal stability primarily due to a strong net positive surface charge. However, the material is highly susceptible to stress-induced aggregation when subjected to external mechanical energy. Quantitative modeling using the Fréchet distribution demonstrated that sonication shifts the particle size mode from 2.7 um to 4.0 um, confirming that mechanical stress facilitates the formation of larger, persistent aggregates. This behavior is attributed to the inherent charge heterogeneity of the particles, which allows attractive forces between oppositely charged subpopulations to dominate once the electrical double layer is momentarily disturbed.

The identified baseline particle size range (mode: 2.6 to 2.9 um) is highly favorable for infiltration into the micro-voids of fibrous preforms used in radio wave absorption composites. To ensure successful fabrication and uniform integration, it is critical to employ low-shear and low-energy infiltration techniques. Maintaining the particle size below the observed aggregation threshold is essential for achieving deep, uniform infiltration and optimizing the functional electromagnetic performance of the resulting composite materials.

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