

## Article

# Classical and modern interpretation of nanofluid stability

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**Abstract:** In this work, the relationships between DLVO theory and dynamic light scattering (DLS) were investigated for a Fe-Cr-Ni-Mn-Si-Mo-Cu nanofluid system obtained through electro-erosive dispersion under the influence of 30 kHz electrical pulses. The increase in hydrodynamic diameter up to 8  $\mu\text{m}$  in DLS measurements proves that attractive forces ( $V_A$ ) outweigh repulsive forces ( $V_R$ ), while the autocorrelation function demonstrates a sharp deceleration of Brownian motion and a total loss of stability. Although the low charge magnitude ( $\zeta < 15 \text{ mV}$ ) and high ion concentration indicate a reduction in the Debye length, there is a possibility of restoring the nano-scale characteristics of these agglomerates through ultrasonic treatment.

**Keyword:** Nanofluid, electrical discharge dispersion, dynamic light scattering (DLS), colloidal stability, DLVO theory.

## Introduction

The DLVO theory (Derjaguin, Landau, Verwey, and Overbeek) serves as the fundamental framework for explaining the stability of colloidal systems. A review of the literature indicates that the interaction between nanoparticles is determined by the sum of two opposing forces: Van der Waals attraction ( $V_A$ ) [1,2] and electrostatic repulsion ( $V_R$ ) energies [3]. Research highlights that the formation of an electrical double layer (EDL) on the surface of nanoparticles creates an energy barrier that prevents particles from approaching one another [4]. Numerous authors have quantitatively demonstrated that increasing electrolyte concentration shortens the Debye screening length (-1), thereby inducing system coagulation [5]. The Dynamic Light Scattering (DLS) method enables the measurement of the hydrodynamic diameter of nanoparticles based on their Brownian motion in a liquid. Literature [6,7] notes that the diameter obtained via DLS is consistently larger than the geometric size measured by transmission electron microscopy (TEM). This discrepancy is explained by the presence of a solvation layer and the ionic atmosphere surrounding the particle. System homogeneity is evaluated through the polydispersity index (PDI) [8,9]. A PDI value below 0,1 indicates a monodisperse system, while a value exceeding 0,3 suggests the onset of agglomeration processes [10,11]. In scientific studies, DLS analysis is widely utilized for the practical validation of DLVO theory [12]. This correlation is significant for several reasons. Using DLS, researchers have monitored changes in particle size across various salt concentrations to identify the Critical Coagulation Concentration (CCC) point predicted by DLVO. The  $\zeta$  potential, measured alongside DLS, serves as a practical measure of the  $V_R$  component in DLVO [13,14]. A value of  $|\zeta| > 30 \text{ mV}$  is generally accepted as an indicator of a high energy barrier and system stability. Contemporary literature [15] emphasizes that classical DLVO theory cannot always fully account for experimental results. Particularly in multicomponent systems, the following additional factors play a role: Steric repulsion: A mechanical barrier formed by molecules adsorbed onto the surface. Hydration forces: Short-range repulsion due to a rigid layer of water molecules. The literature review shows that while DLVO theory remains central to predicting the stability of nanofluids, DLS analysis is essential for verifying this theory under dynamic conditions. However, the relationship between DLVO and DLS in systems subjected to high-frequency electrical pulses (such as 30 kHz) has not yet been sufficiently explored. This article aims to fill this gap by analyzing the variation of DLVO forces under external dynamic energy using DLS data.

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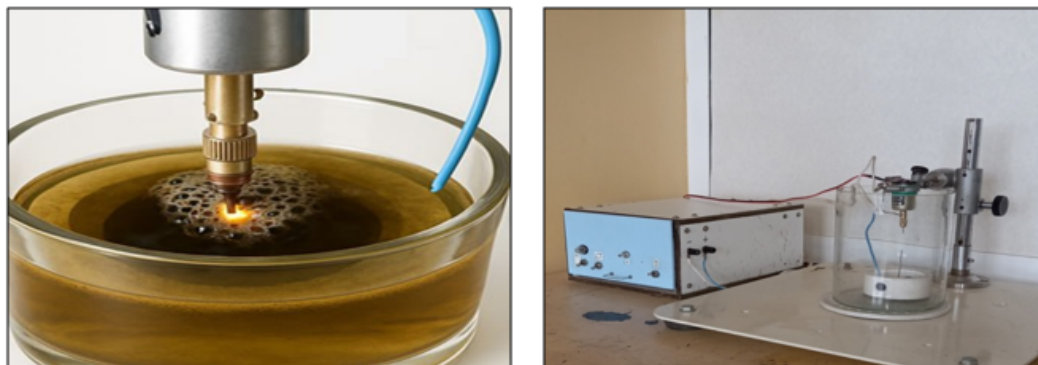
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## Materials and Methods

The dispersion process of the alloy-based sample powders (Figure 1(a)) was conducted using the electro-erosive dispersion device shown in Figure 1(b). The device utilizes a 30 kHz high-frequency power source constructed with high-speed switching IGBT keys, an IRG4PH50UD diode bridge, and 150EBU04 high-voltage diodes. The system comprises a control generator, power supply unit, working head, and a master control system. It delivers a voltage amplitude of up to 450 V and a current of up to 20 A, ensuring high current density for small cathode surface areas.



**Figure 1.** (a) Dispersion process and (b) device illustration

To generate a stable spark discharge, the anode was fully submerged in the electrolyte. A coaxially aligned, vertical conical cathode was immersed into the NaCl aqueous electrolyte medium at a depth of 1–5 mm. Rods made of the sample alloy with a diameter of 2 mm were employed as electrodes. The inter-electrode distance was initially set at 3 mm.

The electrolyte was prepared using chemically pure (CP) NaCl at a concentration of 10 g/L in deionized water with a resistivity of 18 MΩ·cm. The specific electrical conductivity of the electrolyte was recorded as 19 mS/cm at a temperature of 25 °C and a pressure of 101,3 kPa. The hydrodynamic size distribution and dispersion of the nanoparticle agglomerates in the liquid phase were determined using Dynamic Light Scattering (DLS).

## Results

The fundamental premise of the DLVO theory is that the interaction between two nanoparticles in a liquid is the sum of two opposing forces. First, Van der Waals attraction forces ( $V_A$ ) act between particles, tending to make them adhere to one another and form agglomerates. Second, the charges on the particle surfaces create an Electrical Double Layer (EDL), resulting in electrostatic repulsion between similarly charged particles, which maintains the stability of the system by preventing agglomeration.

According to the DLVO theory, the total potential energy ( $V_{total}$ ) is defined by the following formula:

$$V_{total} = V_A + V_R$$

Based on this formula, if particles approach each other very closely, the attractive forces dominate, leading to irreversible adhesion and coagulation. If the repulsive force is sufficient, an energy barrier (or "wall") is formed, preventing the particles from approaching. The higher this barrier, the more stable the nanofluid. Additionally, particles may aggregate loosely at longer distances; such flocculation processes can be reversed through effective stirring.

When an electrolyte like NaCl is added to the liquid, the ions "shield" the surface charges of the particles. Consequently, the Debye length decreases, the repulsive force diminishes, the energy barrier lowers, and the particles precipitate.

If the  $\zeta$  potential ( $\zeta$ ) of the particles is high (typically  $|\zeta| > 30$  mV), the electrostatic repulsion is strong, and the system is considered stable according to DLVO theory. Furthermore, while smaller particles experience weaker Van der Waals attraction, this does not always guarantee stability.

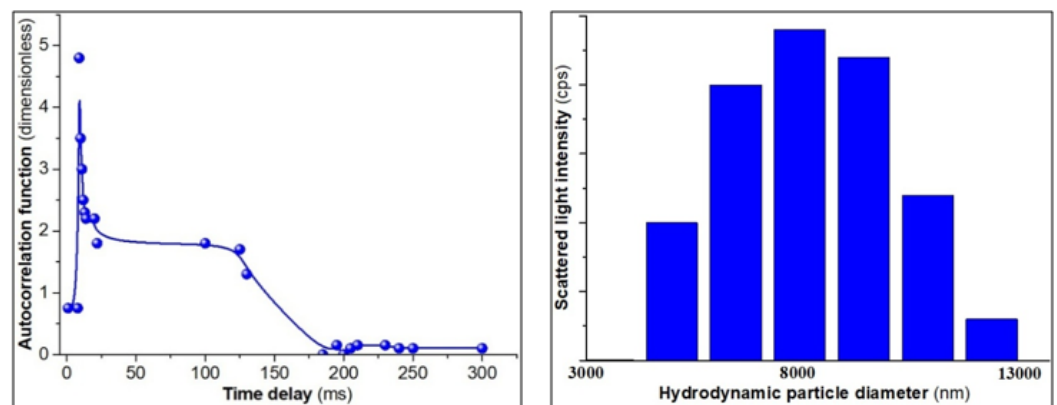
The correlation between the DLVO theory (the stability model) and the DLS measurement method clearly expresses the "theory and practice" relationship in nanofluid research. The DLS instrument is the primary tool for quantitatively confirming the changes predicted by DLVO theory. Their relationship can be observed in three key points:

$\zeta$  Potential Measurement: DLS instruments typically measure the  $\zeta$  potential. According to DLVO theory, a  $\zeta$ -potential higher than 30 mV indicates a high energy barrier and a stable system. Thus, the  $\zeta$ -potential value obtained via DLS provides a practical demonstration of theoretical stability.

Particle Size Monitoring: According to DLVO, if attraction forces ( $V_A$ ) exceed repulsive forces ( $V_R$ ), particles adhere and grow in size. DLS measures the hydrodynamic diameter of these particles. If the diameter measured by DLS increases over time, it signifies that the energy barrier has been breached and particles are falling into the "primary minimum."

Critical Coagulation Concentration (CCC): The CCC is the most important practical application of the DLVO theory. It allows for the prediction of when a system will destabilize upon the addition of an electrolyte. In experiments, NaCl is added incrementally, and the particle size is measured via DLS at each step. At a specific concentration (the CCC point), the particle size begins to increase sharply. This point practically proves the drastic reduction of the "Debye screening length" and the reduction of the energy barrier to zero.

The following are the DLS results for the Fe-Cr-Ni-Mn-Si-Mo-Cu system, obtained by dispersing a multi-component alloy in an electrolyte medium using electro-erosion (refer to Figure 1).



**Figure 2.** (a) Autocorrelation function, (b) Hydrodynamic diameter

The presented graphs are the results of DLS (Dynamic Light Scattering) analysis, and they provide very important information about the stability of the nanofluid. We analyze this based on DLVO theory. In the graph in Figure 3(b), it can be seen that the hydrodynamic diameter of the particles is from 3000 nm to 13000 nm, i.e., from 3 to 13 microns. The central peak is around approximately 8000 nm (8  $\mu$ m). For nanoparticles, this is an extremely large size. If the size of the nanoparticles in the Fe-Cr-Ni-Mn-Si-Mo-Cu system used as a sample was 20-50 nm in TEM analysis [ ], its appearance as 8000 nm in DLS means that the particles have undergone full agglomeration (sticking). So, the attraction forces ( $V_A$ ) in DLVO theory prevailed over the repulsion forces ( $V_R$ ). The energy barrier disappeared and the particles fell into the depth of the "primary minimum".

In Figure 3(a) above, the decrease of light signals over time is shown. The autocorrelation curve extends to 125-175 ms and then drops to zero. In stable nanofluids, this decrease should have happened within microseconds ( $\mu$ s). The larger the particles, the slower their Brownian motion. The slowing down to 175 ms in the graph confirms the presence of very heavy, low-mobility agglomerates in the system. This graph is kinetic proof showing that the system has passed the "critical coagulation" stage and that colloidal stability is completely lost. According to this analysis, the sample nanofluid is in an unstable state. The reasons for this may be the following. The charge on the surface of the particles is not sufficient ( $|\zeta| < 15$  mV), as a result, electrostatic repulsion cannot overcome Van der Waals attraction. The concentration of ions in the liquid is high, which shortened the Debye length and destroyed the energy barrier. After the 30 kHz vibration of the dispersion device stopped, the

balance of DLVO forces was disturbed, and the particles immediately stuck together. However, the state of sticking, i.e., being agglomerated, does not exclude the possibility of breaking the nanoparticle agglomerate again under the influence of ultrasound when reused.

### Discussion:

Dynamic Light Scattering (DLS) analysis results show the colloidal instability and strong agglomeration of the nanofluid in the Fe-Cr-Ni-Mn-Si-Mo-Cu system. Despite the particle size being 20–50 nm in TEM analysis, the hydrodynamic diameter reaching up to 8000 nm (8  $\mu\text{m}$ ) in DLS confirms that according to DLVO theory, attraction forces ( $V_A$ ) prevailed over repulsion forces ( $V_R$ ) and the particles overcame the energy barrier and fell into the depth of the "primary minimum". The extension of the autocorrelation curve to 175 ms kinetically proves that Brownian motion has slowed down sharply and the system has passed the "critical coagulation" stage, completely losing stability. Although the low amount of charge ( $\zeta < 15 \text{ mV}$ ) and high concentration of ions show the shortening of the Debye length in this case, there is a possibility of re-decomposing these agglomerates with ultrasound and temporarily restoring the nano-sized properties of the system.

### Authors' contribution

Conceptualization, A.Z. and B.G.; methodology, A.Z.; validation, formal analysis, investigation, data curation, writing the original draft, writing and editing, project administration. All authors have read and approved the published version of the manuscript.

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### Data Availability Statement

No new data were created and data is unavailable due to privacy

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The authors declare no conflict of interest. Sponsors did not participate in the development of the research; in the collection, analysis, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

### Abbreviations

The following abbreviations are used in this manuscript:

CCC	Critical Coagulation Concentration
CP	Chemically pure
DLVO	Derjaguin, Landau, Verwey, and Overbeek theory
DLS	Dynamic Light Scattering
EDL	Electrical Double Layer

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