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Original Research Paper

Ultrasound-assisted synthesis of colloidal nanosilica from silica fume: Effect of sonication time on the properties of product

Vajihe Jafari, Ali Allahverdi *, Mostafa Vafaei

Cement Research Center, School of Chemical Engineering, Iran University of Science and Technology, Narmak 16846-13114, Tehran, Iran

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ABSTRACT

Current methods of colloidal nanosilica production are relatively energy-intensive and in some cases not environmentally friendly and therefore essential needs are felt to develop new low cost environmentally friendly methods. This study is devoted to the synthesis and characterization of colloidal silica nanoparticles prepared from silica fume using ultrasound. Colloidal nanosilica has been synthesized via dissolution-precipitation process followed by applying ultrasonic waves with the power and frequency of 30 W and 20 kHz, respectively. The produced colloidal nanosilica was characterized via dynamic light scattering (DLS), scanning electron microscopy (SEM), transmission electron microscopy (TEM), Fourier transform infrared (FTIR) spectroscopy and measurements of its zeta potential and specific surface area. DLS results show that minimum particle size, average diameter and maximum particle size of the produced colloidal nanosilica decrease sharply from 28.21, 54.92 and 164.20 nm to 18.17, 38.72 and 141.80 nm, respectively, during the first 15 s of sonication. No significant changes have been observed in applying continued sonication up to 60 min. Measurements of zeta potential confirmed a relatively good stability of the produced colloidal nanosilica.

1. Introduction

Nanosilica, one of the most practical nanoparticles, has been used in various industrial applications such as reinforcing agents, pigments, pharmacy, thermal and electrical insulators, electronic and mechanical materials as it has new physicochemical properties which do not appear in the corresponding bulk materials [1]. Also, over periods of time, many other applications have been found for it. The main problem of silica nanoparticles in powder form is the extreme agglomeration of nanoparticles during the drying process. This problem may be solved by applying some costly or difficult techniques such as adding surfactants and modifying the drying methods. For this reason, production of colloidal nanosilica and characterization of its properties are attaining more and more importance.

Colloidal nanosilica is referred to a liquid medium such as water in which silica nanoparticles are dispersed as dispersed phase. Colloidal silica can be produced through several methods such as neutralizing soluble silicates with acids, dialysis and electrodialysis, dispersion of pyrogenic silica [2], ion exchange [3–6], peptization [7], hydrolysis of silicon compounds [8–10] and dissolution of elemental silicon [11–13]. The main differences between these methods are based on the starting source material, particle size and shape, stability, cost and energy consumption [14].

Ion exchange method, among other methods, is generally used to synthesize colloidal nanosilica. Despite its relatively easy control of particle size, it has some disadvantages including the presence of sodium ions in the product, recycling of ion exchange resin and its environmental costs. However, other production methods have limitations such as using high cost raw materials like tetraethyl orthosilicate (TEOS), recycling of solvent, energy consumption, size and shape limitation [14]. Therefore, considering the growing needs, the resulting product is not cost-effective and it is necessary to find alternative cost-effective production methods.

On the other hand, recent research activities have confirmed the capability of ultrasonication technique for synthesis and production of some nanoparticles [15–18]. The basis of ultrasonication technique, which is currently being developed in various fields of industrial and medical as well as in chemistry and chemical synthesis of nanoparticles, is acoustic cavitation phenomenon involving the formation, growth and collapse of bubbles in a solution. In this technique, the ultrasound energy is transferred to the solution by wave motion, and the compression and rarefaction cycles of

* Corresponding author. Tel.: +98 21 7724 0475; fax: +98 21 7724 0397.
E-mail address: ali.allahverdi@iust.ac.ir (A. Allahverdi).

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ultrasound waves result in bubble formation. The bubbles then grow fast and reach their unstable size. The energy released as a result of the collapse of bubbles can lead to chemical and mechanical effects [19].

The formation of shock waves associated with the collapse of bubbles occurs during the ultrasonic irradiation of solution. The shock waves created by acoustic cavitation increase the momentum of particles and cause them to collide into each other with large force [20]. The driving force for these collisions is the eddies induced by cavitation phenomenon in which the particle breakage occurs due to collisions with other particles, the ultrasonic horn tip, the wall of vessel and eddies [21]. The collisions among particles lead to agglomeration of particles and the agglomerated particles are thus eroded and split [20–24]. Mechanical breakage of the suspended particles induced by shock waves created by cavitation phenomenon during ultrasonic irradiation of suspension is related to the power and frequency of the ultrasound waves, the physical properties of the medium, etc. [21]. In the erosion mechanism single particles are separated one after another from the fracture surface whereas in the fragmentation mechanism fractures are split into smaller agglomerates [22]. The models for fragmentation and erosion of particles exist in literature as Aoki et al. [25] introduced a simple model for fragmentation of particles in an ultrasonic dispersion process. The model is based on the cluster breakage during ultrasonication by interaction of the agglomerates with collapsing cavities in the liquid [26].

As mentioned previously, a disadvantage of current production methods of nanosilica is the use of relatively high cost source materials. Alternative low cost source materials can significantly reduce the production expenses. One of such materials is silica fume which is sometimes referred to as microsilica. It is a by-product of silicon metal and ferrosilicon alloy industries that possesses a high content of amorphous silica (more than 85 wt.%) and a high level of specific surface area. Silica fume can be considered as an all-purpose mineral which has been used in different industries like cement and concrete [27].

Lee et al. [7] prepared colloidal silica via peptization method, but there is no report in the literature for the application of silica fume and ultrasonication technique together in peptization method.

The current study is devoted to evaluating the potential of ultrasonication technique and the suitability of silica fume as the source material for the synthesis of colloidal nanosilica. Affirmative outcomes in this regard are invaluable innovations as they can provide opportunities for replacing autoclave operation in peptization method with ultrasonication technique which can effectively reduce the time and energy consumption in converting silica gel to colloidal nanosilica. It should be noted that the influencing parameters on the properties of produced colloidal nanosilica are many. They include parameters related to ultrasonication conditions affecting cavitation phenomenon such as power, frequency of the ultrasound irradiation, sonication time, ultrasonic intensity and also those related to the synthesis conditions such as reaction time, reaction temperature, pH and reagents concentrations. The present work, however, does not cover all these influencing parameters, except the sonication time and its influence on particle size distribution of produced colloidal nanosilica. Extensive studies are required to evaluate the effects of all these parameters. These studies are necessary for the purpose of optimizing the synthesis conditions.

To obtain colloidal silica nanoparticles by the application of ultrasound, the experiments have been done at relatively low power and frequency of the ultrasound irradiation of 30 W and 20 kHz, respectively. This is because at very high frequency, the rarefaction and compression cycles are very short which is insufficient to initiate cavitation so that chemical and mechanical effects of ultrasound irradiation will be less [19,28]. When the power of ultrasound waves increases, the formation of cavitation bubble in solution increases which it blocks the ultrasound energy transfer from the probe to the solution [28].

2. Experimental work

2.1. Materials and apparatus

Silica fume, sodium hydroxide pellets (97%, Merck & Co.), sulfuric acid (Analytical grade, 95–97%, Merck & Co.), hydrochloric acid fuming (Analytical grade, 37%, Merck & Co.) and extra pure crystalline sodium chloride (99.5%, Merck & Co.) have been used in this study. The silica fume was provided by Iran Ferroalloy Industries Company. The chemical composition and physical properties of the silica fume are given in Table 1. As can be seen, with a silica content of almost 96%, it can be considered as a relatively highly pure source material.

A Bandelin ultrasonic probe system, HD 3200 with tapered tip KE 76 made of titanium alloy was used for the ultrasonic irradiation. The power of the apparatus was maintained constant at 30 W and the frequency of the waves was 20 kHz.

2.2. Method

The procedure used to synthesize colloidal nanosilica from silica fume is depicted schematically in the diagram shown in Fig. 1. At first, 1 g of the silica fume was mixed with 100 ml 2.5 M sodium hydroxide aqueous solution and then the mixture was heated up to about 80 °C for 30 min with continuous constant stirring. According to Eq. (1), the silica was converted to sodium silicate solution. Subsequently, the suspension was filtered through a filter paper of Whatman grade 41 to remove the insoluble solids. After filtration, the obtained sodium silicate solution was stirred with a continuous constant rate and neutralized with 2.5 M sulfuric acid aqueous solution until pH was adjusted in the range of 8–8.5. Eq. (2) shows the corresponding chemical reaction in this step. The formed silica gel was then washed with distilled water for 5–6 times to remove solute salts like sodium sulfate. After washing, the diluted sodium hydroxide aqueous solution was added to the precipitated silica gel under constant stirring to adjust pH in the range of 8.5–10.5. The produced silica gel was then converted to colloidal nanosilica by applying ultrasonic waves with the power and frequency of 30 W and 20 kHz, respectively, in periods varying from 15 s to 60 min.

$$SiO_2 + 2NaOH \rightarrow Na_2SiO_3 + H_2O \tag{1}$$

$$Na_2SiO_3 \rightarrow H_2SO_4 \rightarrow SiO_2 + Na_2SO_4 + H_2O \tag{2}$$

2.3. Characterization

Particle size distribution and zeta potential measurements, scanning and transmission electron microscopy (SEM & TEM)

Table 1
Chemical composition and physical properties of the silica fume.

Component	Value
SiO_2 (%)	96.12
Al_2O_3 (%)	0.82
K_2O (%)	0.4
LOI (%)	0.63
Density (g/cm^3)	0.213
Specific surface area (m^2/kg)	18,000

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