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Abstract. This studied focusing on the effect of surfactant, pH, and viscosity to produce stable silica nanofluid. For comparison we synthesize silica nanofluid with the addition of surfactant sodium lauryl sulfate. Silica nanofluid was synthesized by one step synthesis with the help of microbubble from microbubble fine generator. In the microbubble fine generator CO_2 gas and sodium silica solution will contact and carbonation process occurs, then the solution will enter the reactor to increase the contact time of sodium silica and CO_2. The final product is an overflow solution from reactor and will be characterized by stable nanofluid parameters. From the physical appearance silica nanofluid still transparent. Particle size and zeta potential are two important factors to consider in evaluating the stability of the silica nanofluid. Result indicates that the stability of silica nanofluid highly depends on the presence of surfactant. On the basis of these experiments, the silica-surfactant nanofluid has been successfully obtained with the zeta potential and particle size both are -53.233 mV and 5.616 nm. While for silica nanofluid has zeta potential and particle are -24.2 mV and 4.849 nm. From the results we can obtained that the addition of sodium lauryl sulfate in the synthesis of silica nanofluids can increase the stability.

Keywords: silica nanofluid, sodium lauryl sulfate, carbonation, microbubble.

INTRODUCTION

Nanofluids are a new class of fluids engineered by dispersing nanometer-sized in base fluids. They are two-phase systems with one phase (solid phase) in another (liquid phase). Nanofluids have been found to possess enhanced thermophysical properties such as thermal conductivity, thermal diffusivity, viscosity, and convective heat transfer coefficients compared to those of base fluids like oil or water. It has demonstrated great potential applications in many fields. One kinds of nanofluids that find many technological potential applications are silica nanofluid (1).

Silica is the second abundant compounds found in the earth crust. Silica nanoparticles with controlled properties for a specific application are typically prepared from alkoxide compounds, e.g., tetraethyl orthosilicate (TEOS) or tetramethyl orthosilicate (TMOS). However, alkoxides are expensive and toxic that may hinder their applications for commercial scale. Sodium silicate, an inexpensive and abundantly available material, may become an alternative as raw material to prepare advanced functional silica with controlled properties. Sodium silicate is usually made of quartz sand or biogenic silica that can be further converted into silica gels and powders (2). However, the precipitation rate in sodium silicate system is typically fast that causes a difficulty in controlling the particle size.

Silica-based nanofluids have been synthesized using various methods, ranging from commercial silica dispersed directly in certain fluids using ultrasonic waves to direct synthesis of the silica source by the sol-gel method. Silica sources for the synthesis of nanofluids usually use tetraethylortho silicate (TEOS) and sodium silicate (Na_2SiO_3).

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The reaction for the formation of silica nanoparticles from TEOS proceeds more slowly than Na_2SiO_3 so that the morphology and size of the particles dispersed in the fluid is easy to control, but silica derived from TEOS is synthesized in an alcoholic atmosphere using the Stober method because TEOS does not have a hydroxyl group (-OH). In addition to not having hydroxyl groups, TEOS-based silica nanofluids require a binder such as xylene so that the silica nanofluids have good stability. However, the lack of efficiency in the synthesis process and the relatively high price began to shift to using abundant sources of silica such as sodium silicate.

Sodium silicate-based silica nanofluid synthesis has been synthesized using the sol-gel method with ion exchange. The sol-gel method in the synthesis of silica nanofluids has the advantage that the morphology and particle size can be controlled at ambient temperature based on changes in the pH value. The most common method to prepare silica nanofluids is by dispersing nano-sized silica particles into base liquid. To enhance the stability of the suspension from sedimentation and aggregation, surfactant is usually added (3) (4) (5). Qomariyah et al. (6) prepared a stable colloidal silica from sodium silicate by adding surfactant and KOH. The colloidal silica was used as precursor for spray dryer to produce silica particles. Cai et al. (8)using carbonation process with CO_2 gas for the process of making silica precipitates from sodium silicate, and the addition of surfactants to sodium silicate solution.

The purpose of this work is to develop a method to prepare silica surfactant-stabilized nanofluid from sodium silicate by carbonation in a micro-bubble column. The use of micro-bubble in contacting carbon dioxide gas is aimed to significantly increase the mass transfer rate. Effect of carbonation can facilitate the process of synthesis nanofluids. In this work, we report the synthesis of silica nanofluids by carbonation of sodium silicate solution in one step at ambient temperature and low-pressure conditions. The synthesis of nanofluids was carried out with the addition of surfactants as stabilizers. The resulting silica nanofluid will be characterized according to the standard for testing the stability of the nanofluid which includes physical appearance, changes in pH and viscosity values, zeta potential value, and size distribution of silica nanoparticles in the fluid.

MATERIALS AND METHOD

Materials

Sodium silicate (28% SiO_2; SiO_2:Na_2O = 3.3) was provided by PT. PQ Silica Indonesia. An anionic surfactant of sodium lauryl sulfate (SLS; reagent grade) was purchased from CV. Mitra Sari Kimia. High-purity carbon dioxide gas (99%) was supplied by PT. Samator Gas Industry. Deionized water was purchased from UD. Sumber Ilmiah Persada. All chemicals were used as received without further purification.

Experimental

The schematic diagram of apparatus to synthesize nanofluid is shown in Figure 1. It mainly consisted of: (i) a glass column, (ii) a fine bubble generator, and (iii) a gravity flow tank. The column as the reactor to form stabilized silica nanofluid was made of acrylic with 9.5 cm in diameter and 113.5 cm in height. The fine bubble generator was a mechanical type where CO_2 bubbles injected from pressured gas tank into sodium silicate solution was broken up by a propeller. The carbonation process is carried out at an ambient temperature and the gas flow rate used is 0.7 L/min at a pressure of 1.5 atm.

The sodium silicate solution with a concentration of 0.5% was pumped into the glass column. The 0.5% sodium silicate solution was prepared by diluting the 28% sodium silicate solution with demineralized water. Sodium lauryl sulfate was also added into the solution during its preparation with a 5 critical micelle concentration (CMC), where in 5 liters of 0.5% sodium silicate solution added 1.5 g of SLS. When the solution reaches a certain height, it will flow into the fine bubble generator. CO_2 gas and the solution will come into contact thus the carbonation process occurs. Then, the solution is pumped back into the glass column to prolong the contact time between the solution time and CO_2 gas, where this process will continue until it overflows. The solution flowed out of the gravity tank as a final product, then evaluated with many kind parameters of stable nanofluid, one of them is pH measurement. The process was continued because flow rate of the sodium silicate solution was fixed constant at 100 mL/min.

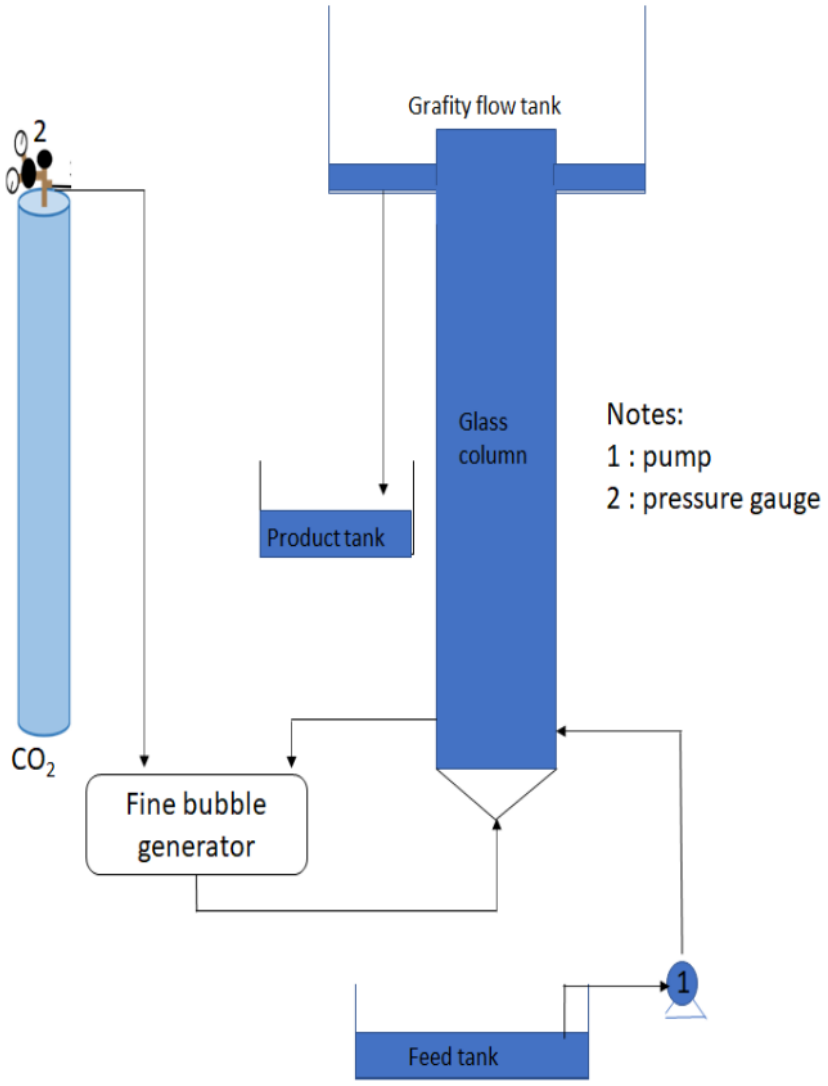

FIGURE 1. *The Schematic Diagram of Microbubble Reactor*.

Characterization

The parameter of important concern in this study is the stability of the silica nanofluid produced from each method. Physical observations and changes in pH value are carried out periodically to observe whether deposits or particle aggregation are formed on silica nanofluids (with Sodium Lauryl Sulfate surfactant or without surfactant). Observations were also made on the zeta potential value and particle size distribution in the silica nanofluids using the Malvern Zeta sizer-ZS instrument. Measurement of the value of the particle size distribution is carried out periodically to observe changes in the size of the silica particles. Changes in the viscosity of the nanofluids were also observed using the Ostwald viscometer to identify the type of silica particles growth in the nanofluids

RESULTS AND DISCUSSION

RESULTS

Figure 2 shows photographs of silica nanofluids and silica silica-surfactant nanofluid at different sedimentation time. Initially, the color clear (fig. 2a and 2b). After a month (fig. 2c), there is no sediment can be observed. It indicates that silica nanofluid and silica-surfactant nanofluid are still stable. Apart from physical appearance, pH is also one of the stability parameters of nanofluids. The pH level is an important factor affecting the stability of nanofluids. In this study, a surfactant Sodium Lauryl Sulfate was used as a dispersing agent to obtain transparent and stable nanofluids. As a comparison, pH of the nanofluid without surfactant was measured

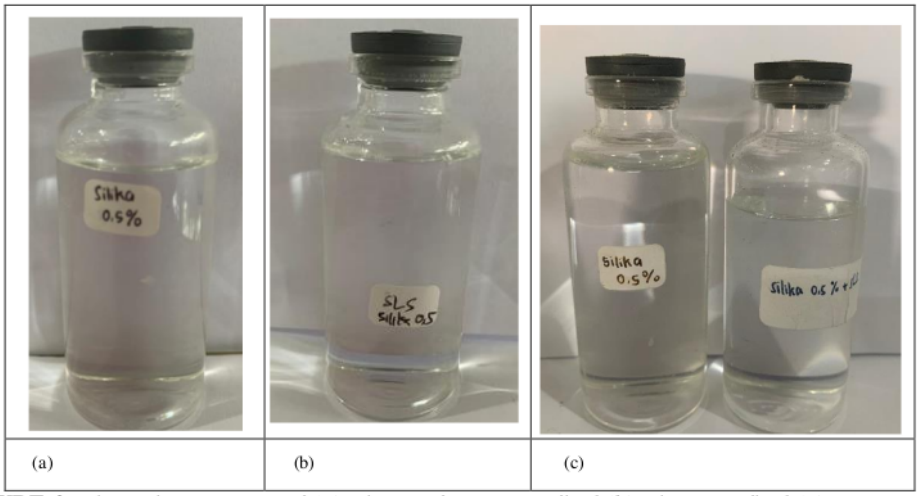

FIGURE 2. *Physical appearance of (a) silica-surfactant nanofluid (b) silica nanofluid (c) comparison of silica surfactant nanofluid and silica nanofluid for a month.*

Table 1 shows the pH evolution of silica nanofluid with and without the addition of SLS surfactant. The initial pH of nanofluid was 10.538 and 9.346 for, respectively, without surfactant and with surfactant. For both cases, the pH tended to decrease gradually from day-1 to day-5. However, the decrease in pH for nanofluid without surfactant is larger than that with surfactant. For the nanofluid without surfactant, the decrease is approximately 3% and for that with surfactant is only 1%. The lower decrease in pH with time may indicate that the silica nanofluid with surfactant is stable than that of without surfactant. The decrease in pH may be cause by the condensation reaction of surface silanol groups with the remaining ortho silicic acid in water (2):

$Si(OH)_4 + HO–Si(OH)_3 \rightarrow Si(OH)_3–O–Si(OH)_3 + H_2O$. The presence of surfactant in the solvent may hinder the direct contact between the surface silanol groups and the ortho silicic acid. This may be the reason why the pH of silica nanofluid with the presence of surfactant only decrease slightly.

TABLE 1. *Comparison pH of silica-surfactant nanofluid and silica nanofluid for some days*

Silica-surfactant		Silica	
Time	pH	Time	pH
Day-1	9.346	Day-1	10.538
Day-4	9.315	Day-4	10.341
Day-5	9.252	Day-5	10.230

Several parameters to determine the stability of nanofluids beside of from the presence or absence of agglomeration, its zeta potential and particle size. The zeta potential value indicates the magnitude of the repulsion between nanoparticles dispersed in the fluid. The magnitude of the force can state the possibility of aggregation between particles which causes the nanofluid to become unstable. The zeta potential of ± 30 mV is regarded as a critical value for evaluating the stability of the nanofluids. A lower zeta potential (<30 mV) leads to particle agglomeration, whereas a higher zeta potential (>30 mV) can maintain stability (8). The zeta potential of silica nanofluid is -24.2 mV. The zeta potential of ± 30 mV is regarded as a critical value for evaluating the stability of the nanofluids. A lower zeta potential (<30 mV) leads to particle agglomeration, whereas a higher zeta potential (>30 mV) can maintain stability (8). This shows that silica nanofluid less stable because it has potential value less than absolute 30 mV. A low zeta potential value can indicate particle agglomeration, the occurrence of this agglomeration can be seen from the particle size distribution. Figure 3 shows comparison result particle distribution (PSA) of silica nanofluid (1a) and silica-surfactant (1b) for a day. The result particle distribution of silica nanofluid obtained by the highest peak intensity at 4.849 nm, this size can classify as nanofluid because of the size is not more than 100 nm. Because of particle size is small, so that the silica nanoparticles have a low probability of aggregation (4).

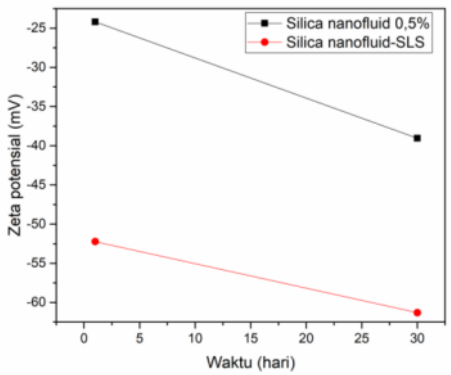

FIGURE 3. *Zeta potential value of silica nanofluid 0,5% and silica-sls nanofluid in a day and after a month*

Otherwise as a comparison to know influence of SLS as a surfactant, same several characterizations on the silica nanofluid were obtained. From fig.3 we know that zeta potential value of silica-sls nanofluid is greater than silica nanofluid, even after a month. The zeta potential value of silica-sls nanofluid on day 1 is -53.233 mV. This shows that the presence of SLS can decrease zeta potential values. It remains that silica nanofluid stable and produces a good dispersion, so that silica nanoparticle can be maintained with the expected size. The low zeta potential value can indicate particle aggregation, the occurrence of this aggregation can be seen from the particle size distribution. Figure 4 shows the particle size distribution (PSA) obtained by the highest peak intensity at 5.616 nm, this small enough size minimized the occurrence of aggregation and form stable silica-surfactant nanofluids.

The presence of SLS also has a function to stabilize colloidal silica through steric stabilization. Since the SLS head groups which are hydrophilic groups form a physical bond to the silica surface, their tail portion which is the hydrophobic group congregates with the hydrophilic group portion of the rest of the surfactant. The free swing of the surfactant tail in the fluid can provide a protective layer that extends into the solution which can form a barrier, in addition to an electrostatic barrier, to seal off particle approaches and minimize aggregation between silica particles. This dual action increases the stability of silica colloidal. Thus, SLS mobilized on the silica surface appears to have the function of increasing colloid stability (7).

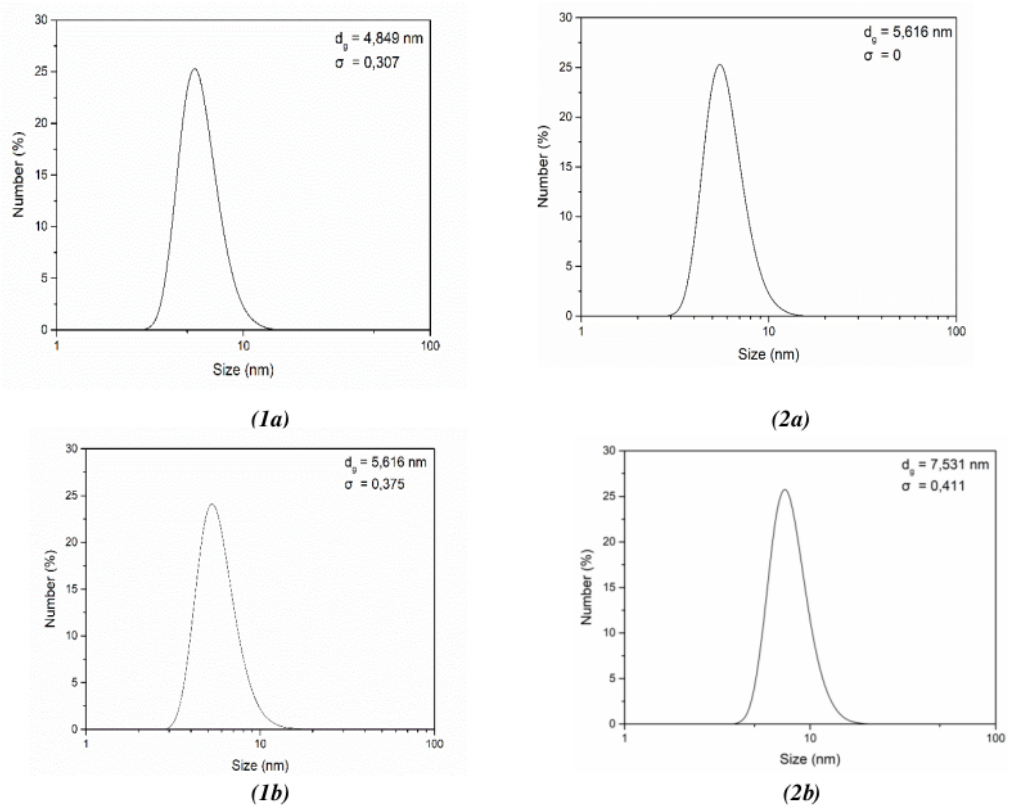

FIGURE 4. *Comparison particle distribution of (a) silica nanofluid 0,5% (b) silica-surfactant in a day and after a month*

After a month the zeta potential values of silica nanofluid and silica-sls nanofluid both are -39.033 mV and -61.3 mV. So from the result, it can be known that silica nanofluid can remains stable for a month, because it have zeta potential more than absolute 30 mV (4). Then, to find out there is no aggregation, distribution particle was measured, Fig.4 shown that particle size (PSA) for silica nanofluid and silica-sls nanofluid both have the highest peak intensity at 5.616 nm and 7.513 nm.

Because from the results measuring of the size distribution particles, the PSA value has increased, this can indicate that the particles are growth by the time. The explanation for this particle growth can be related to the viscosity value. Where in table 2 can show from the viscosity of the stable silica nanofluid for a month. The relationship between viscosity and particle size is that viscosity increases with increasing particle size. Although it seems that particle size influences the viscosity of a nanofluid, the effect of the other issues also should be considered. Nanoparticles can easily agglomerate and make clusters, which make it hard to investigate the pure effect of particle size on viscosity of nanofluids. In other words, particle size of nanoparticles would be increase due to clustering and aggregation of nanoparticles, which consequently leads to an increase in (11). The effect of the addition of surfactants on viscosity, namely that the viscosity value tends to increase because the solution becomes thicker and affects the viscosity value (12).

TABLE 2. *Observation the stability of colloidal silica to time*

Silica-surfactant			Silica		
Time	Viscosity (poise)	Particle size (nm)	Time	Viscosity (poise)	Particle size (nm)
Day 1	0.0921	5.616	Day 1	0.096	4.849
Day 30	0.0933	7.531	Day 30	0.096	5.616

CONCLUSION

One step synthesis of silica nanofluids with the he of microbubbles has been successfully carried out by producing silica nanofluids. The addition of the SLS surfactant increase the stability of the silica nanofluid, it can be known from zeta ential of silica-surfactant nanofluids is -53.333 mV, while the zeta potential values of silica nanofluids is -24.2 mV. The particle size indicates the stability of the silica nanofluid, while particle size for silica nanofluid and silica-surfactant nanofluids both are 4.849 nm and 5.616 nm so, it can be indicated that particle size of silica nanofluid is small and there is no possibility agglomeratio happen. To determine the stability of the nanofluid silica, zeta value and particle size were measured after a month. The zeta potential and particle size values of silica nanofluid and silica-surfactant nanofluid increased. Increase of particle ize indicated that there's particle growth. From the results we can obtained that the addition of sodium lauryl sulfate in the synthesis of silica nanofluids can increase the stability

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