

Effect of Solution Concentration, Fly Ash Ratio, and Aging Time on the Quality of Nano-Silica

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Abstract

Fly ash, a byproduct of coal combustion, has gained significant attention due to its high silica content, and this silica-rich waste can be effectively converted into nano-silica. Factors such as the molarity of the solution, liquid-to-solid ratio, and the aging time play an essential role in determining the characteristics of silica nano produced, including particle size, morphology, and purity. This research aimed to determine the optimal conditions of solution molarity and the liquid-to-solid ratio to produce high-purity nano silica, examine the effect of aging time on the morphology and size of silica nanoparticles, and evaluating the effectiveness of the sol-gel method in producing nano-silica. This research uses nitric acid (HNO₃ 3 M) as the solution because it has not been widely explored and is more effective than other acidic solutions. In addition, the liquid-to-solid ratio varied from 1:5, 1:10, and 1:15, and combined with an aging time of 9 and 12 days, this study became the focus. The sample tested included XRD, XRF, FTIR, and SEM to confirm the nano-silica characteristics. It shows XRD is in the range of 2θ of about 20-22°, which indicates the presence of amorphous silica, and FTIR shows results that support XRD data. SiO₂ composition ranges from 97.47% to 98.57%, a very high amount of pure silica. It may concluded that silica nanoparticles have a particle size that varies from 151.2 nm to 295.4 nm, with a smooth morphology and relatively well distributed.

Keywords

Fly Ash, Nano-Silica, Particle Size

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1. INTRODUCTION

Fly ash is a byproduct of coal combustion that has gained significant attention due to its high silica content (Agi et al., 2020; Alterary and Marei, 2021; Manchanda et al., 2017; Vashistha et al., 2019; Yadav et al., 2019; Yan et al., 2016; Journal et al., 2024). This silica-rich waste can be effectively converted into nano-silica, a nanomaterial characterized by a large surface area and high reactivity (Septriansyah and Saggaff, 2021; Yadav and Fulekar, 2019). Nano-silica can be applied in a wide range of fields, including construction, catalysis, and energy production, underscoring its versatility and potential for industrial use (Kamara et al., 2023; Kola and Usman, 2023; Septriansyah et al., 2025). One commonly used method for synthesizing nano-silica is the sol-gel process, which uses chemical treatment with an acidic or alkaline solution to extract silica from fly ash. Factors such as solution molarity, the solution-to-fly-ash ratio, and aging time significantly influence the properties of silica-nano, such as particle size, shape, and purity (Ge et al.,

2018; Miricioiu and Niculescu, 2020).

The molarity of the solution, whether acidic or alkaline, plays a crucial role in extracting silica from fly ash. A higher molarity can speed up fly ash decomposition and increase silica yield (Miricioiu and Niculescu, 2020; Zhang et al., 2021). However, excessive molarity can cause overwashing, damaging the silica structure. NaOH solutions with the right molarity help produce purer nano-silica with uniform particle sizes (Caroles, 2019).

The solution-to-fly-ash ratio also impacts nano-silica synthesis (Gao et al., 2017; Liu et al., 2018). A low solution ratio reduces silica leaching efficiency, leaving impurities like Al₂O₃ and Fe₂O₃. On the other hand, a high ratio of chemicals and water wastes without improving silica extraction. The optimal ratio, around 7:1, ensures maximum silica extraction with minimal waste (Agi et al., 2020).

This research aimed to: 1. Determine the optimal conditions of solution molarity and the ratio of solution to fly ash to produce high-purity nano-silica. 2. Examine the effect of

aging time on the morphology and size of silica nanoparticles, and 3. Evaluating the effectiveness of the sol-gel method in producing nano-silica. This research is expected to produce nano-silica with high purity and desired amorphous structure and optimize fly ash processing methods as a raw material with high potential in various industrial applications.

Based on literature reviews, most previous investigations still used acid solutions such as H_2SO_4 , HCl, and NaOH alkaline solutions (Mefatih et al., 2023; Yadav et al., 2019) as a solvent with a high concentration of molarity, thereby the synthesis process becomes complex. In this research, only 1 type of acid solution was used, namely nitric acid (HNO_3), with a concentration of 3 molars, which has not been widely explored and is more effective compared to other acidic solutions (Namvar et al., 2021). In addition, the solid-to-liquid ratio varied at 1:5, 1:10, and 1:15 (Yadav et al., 2023) combined with aging times of 9 and 12 days, forming the primary focus of this research. This method is expected to improve nano-silica's production efficiency and quality and provide a more sustainable solution for utilizing fly ash waste.

2. EXPERIMENTAL SECTION

2.1 Materials

The fly ash material was obtained from PT. Sriwidjaja Fertilizer Palembang. During the experimental processes, fly ash was filtered using strainer No. 200 before usage to remove unwanted dirt and other impurities. Nitric Acid (HNO_3) 65% technical grade was used as the solvent. Distilled water was used in all experiments for washing and making solutions.

2.2 Methods

This research used the sol-gel method to synthesize nano-silica from fly ash with a nitric acid (HNO_3) solution. The acidic solution serves as a solvent to remove impurities like heavy metals (Fe_2O_3 , Al_2O_3) and calcium oxide (CaO) from the fly ash. Key stages in the process include adjusting the solution concentration, the solution-to-fly-ash ratio, and the ageing time. Table 1 outlines the sample code and information.

Table 1. Sample Code and Information

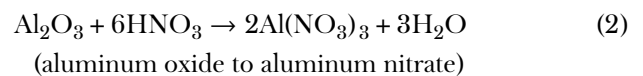
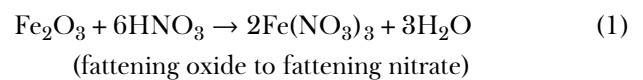
Sample code	Information
3M-RL01-A01	3 Molar, 1:5 ratio, Aging 9 days
3M-RL01-A02	3 Molar, 1:5 ratio, Aging 12 days
3M-RL02-A01	3 Molar, 1:10 ratio, Aging 9 days
3M-RL02-A02	3 Molar, 1:10 ratio, Aging 12 days
3M-RL03-A01	3 Molar, 1:15 ratio, Aging 9 days
3M-RL03-A02	3 Molar, 1:15 ratio, Aging 12 days

This research used three solutions to fly ash ratios such as 1:5, 1:10, and 1:15 (Amin et al., 2016; Yadav and Fulekar, 2019). A higher solution ratio was found to improve the silica dissolution process (Azat et al., 2019). After dissolution, the silica gel was aged 9 and 12 days to enhance silica condensation. Longer curing times were expected to produce nano-silica with

smaller particle sizes and a stable amorphous structure. Finally, the nano-silica was dried and analyzed using XRD and SEM to determine its size, shape, and amorphous properties.

2.3 Acid Treatment

The sol-gel method using HNO_3 3 M solution for the synthesis of nano-silica from fly ash involves several critical stages. Fly ash is mixed with HNO_3 solution with a concentration of 3 M; this reaction is carried out using a magnetic stirrer at room temperature of 25°C for 15 minutes. Equation 1 and 2 shows that Nitric acid reacted with metal impurities such as Fe_2O_3 , Al_2O_3 , and CaO in fly ash, forming water-soluble metal salts, and this reduces the metal levels in the fly ash, leaving behind a purer silica.



In Equation 3, the sodium silicate solution is titrated with HNO_3 slowly until it reaches a pH of about 1-2. Under this condition, silica will settle as silica gel ($Si(OH)_4$). During titration, silanol (Si-OH) groups are formed, forming silica (Si-O-Si) tissues through condensation. This results in an amorphous gel structure.



The silica gel was allowed to age for a predetermined period to stabilize its structure. Subsequently, the gel was washed thoroughly until a neutral pH was achieved and then dried at 100°C to remove residual water from the silica nanoparticles. To improve the silica purity value, the calcination process was also conducted at 600°C (Namvar et al., 2021). Finally, the nano-silica produced through the synthesis process was characterized using XRD tests (X-ray diffraction) and SEM (scanning electron microscopy) to analyze its size, morphology, and amorphous properties. The flow chart of nano-silica synthesis is shown in Figure 1.

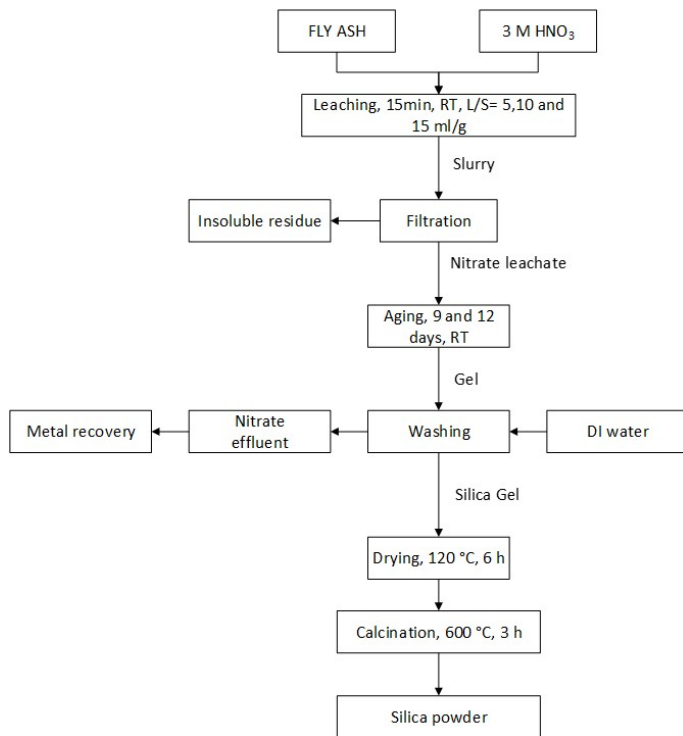
3. RESULTS AND DISCUSSION

3.1 XRD Test Results

The test result of XRD of each nano-silica sample with varying molasses of the solution can be seen in Figure 2. Based on the figure, it can be observed that the majority of samples show major peaks in the range of $2\theta = 20-22^\circ$ indicating the presence of amorphous silica, while the absence of other peaks correlates with the purity value of the extracted silica (Mefatih et al., 2023; Rafryanto et al., 2024). The absence of sharp peaks

Table 2. Chemical Composition of Fly Ash and Nano-Silica (%)

	XRF analysis results (%)								
	SiO ₂	Fe ₂ O ₃	CaO	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	MgO	Na ₂ O	K ₂ O
Fly ash	57.21	5.10	2.75	0.83	29.42	-	-	-	-
3M-RL01-A01	97.65	0.18	0.49	0.4	0.77	0.01	0.16	0.32	<0.01
3M-RL01-A02	97.46	0.10	0.94	0.12	0.74	0.30	0	0.30	<0.01
3M-RL02-A01	98.30	0.10	0.35	0.14	0.73	0	0.08	0.28	<0.01
3M-RL02-A02	98.56	0.08	0.43	0.10	0.55	0.01	0.08	0.16	<0.01
3M-RL03-A01	97.87	0.11	0.77	0.14	0.59	0	0.21	0.29	<0.01
3M-RL03-A02	97.47	0.28	0.65	0.46	0.84	0.01	0.16	0.17	<0.01

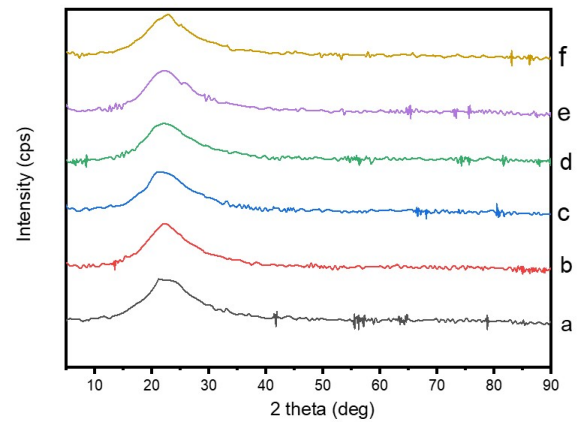
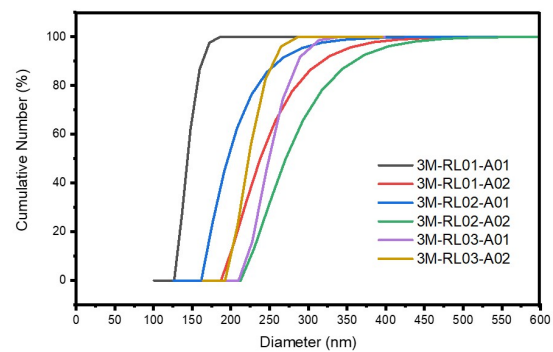
**Figure 1.** Flow Chart of Fly Ash Synthesis

also indicates the absence of an orderly form of crystalline silica confirming the amorphous properties (Imoisili and Jen, 2023).

The absence of sharp crests shows the absence of regular forms of crystalline silica, which confirms its amorphous properties. Additionally, the peak at 22° indicates silica has a uniform morphology (Agi et al., 2020). In other words, this research found that there is a significant influence of solution molarity, the ratio of solution to fly ash, and the selected synthesis technique namely the sol-gel method on the purity of nano-silica.

3.2 XRF Test Results

Table 2 shows the results of the analysis of XRF (X-ray fluorescence) for fly ash and 6 nano-silica samples extracted. The silica content in fly ash is 57.21% which affects the purity of nano-silica made of fly ash. The percentage of SiO₂ in nano-

**Figure 2.** XRD Test Results: a) Sample 3M-RL01-A01; b) Sample 3M-RL01-A02 c). Sample 3M-RL02-A01; d) Sample 3M-RL02-A02; e) Sample 3M-RL03-A01f) Sample 3M-RL03-A02**Figure 3.** Particle Size Distribution Curve

silica samples increased significantly, ranging from 97.47% to 98.56%, and this result is similar to research by Imoisili et al. (2022). Table 2 also confirms that the selected operating conditions and silica fabrication methods through sol-gel could result

in nano-silica with high purity as those components also effectively reduced the percentage of other oxides such as Fe_2O_3 , CaO , and Al_2O_3 to very low levels, or even not detected at all in some samples.

3.3 Particle Size Analysis (PSA)

Particle Size Analysis (PSA) testing is conducted to determine the particle size distribution of materials. Smaller particle sizes can alter the properties of the resulting structure, enhancing its overall strength. The results of the distribution of silica nanoparticles can be seen in Table 3 and Figure 3. The PSA test results with variations in nano-silica samples showed a size of 151.2 nm – 295.4 nm. The 3M-RL01-A01 sample has a silica size of 151.2 nm, while 3M-RL02-A02 has the largest silica size of 295.4 nm. The polydispersity index (PDI) describes the particle size distribution in a sample. Lower PDI values indicate a more uniform size distribution, while higher values indicate a more varied distribution. Table 3 shows that the 3M-RL02-A01 sample had the highest polydispersity index (1.187), which means a highly variable particle size distribution.

Table 3. Particle Size Analysis (PSA)

Sample code	Silica size (nm)	Polydispersity index
3M-RL01-A01	151.2	0.385
3M-RL01-A02	260.1	0.698
3M-RL02-A01	222.4	1.187
3M-RL02-A02	295.4	0.460
3M-RL03-A01	262.5	0.465
3M-RL03-A02	165.0	0.464

In contrast, the 3M-RL01-A01 sample had the lowest polydispersity index (0.385), signaling a more uniform size distribution. It is also considered that the polydispersity value is related to factors such as the decrease in particle size and the molarity ratio. From this point of view, the strong connection between the morphology and size of silica nanoparticles and various values of the polydispersity index can be noticed.

The particle diameter and particle size distribution of silica nanoparticles were determined using a particle size analyzer model from Delsa™ Nano. Figure 3 displays the particle size distribution of the six nano silica samples. Sample 3M-RL01-A01 shows particle size values ranging from 136.7 - 217 nm. The particle size of Sample 3M-RL01-A02 is 202.7 - 572.5 nm. Sample 3M-RL02-A01 shows a range of particle size values from 175.9 - 483 nm, while the particle size of Sample 3M-RL02-A02 is 230.2 - 604.4 nm. Sample 3M-RL03-A01 shows a range of particle size values from 227.4 - 333 nm. Meanwhile, 3M-RL03-A02 shows a range of particle size from 208.4 to 286.9 nm.

Based on Figure 3, sample 3M-RL03-A01 shows the most uniform distribution. This condition can be seen from the sharp curves in specific size ranges. This steepness indicates that most of the particles are very close in size.

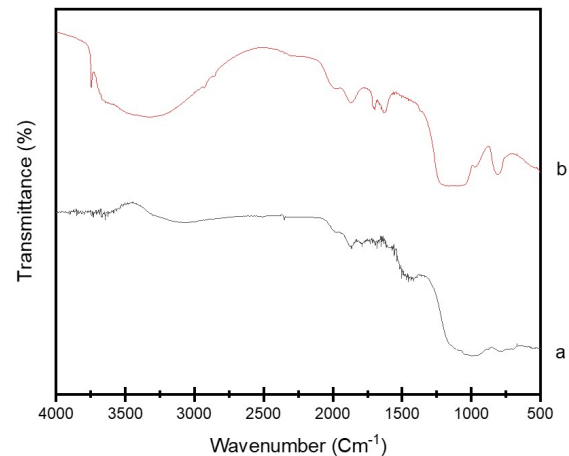


Figure 4. FT-IR Spectra of a. Fly Ash; b. Nano-Silica

3.4 FTIR Test Results

The Fourier transform infrared (FTIR) spectrum of the extracted silica is shown in Figure 4. The FTIR analysis of the sample was recorded using a spectrometer where the spectrum was in the range of 500-4000 cm^{-1} . According to Figure 4, the absence of peaks between 1700 and 2900 cm^{-1} indicates the absence of other organic compounds, as no surfactants or templates are used during extraction. The structural characterization using FTIR shows results that support XRD data. The FTIR spectrum of nano-silica fly ash shows functional groups at wave numbers 3203.76, 1068.56, 975.08, and 810.1 cm^{-1} (Journal et al., 2024).

The Si-OH groups are produced from water molecules absorbed on the surface of silica. The ribbon at 1068 to 1200 cm^{-1} was possibly caused by asymmetrical stretching vibrations from siloxane bonds (Si-O-Si), while the band is at 810 - 975 cm^{-1} assigned to the vibration of the Si-O-Si symmetrical bond stretch. The FTIR analysis results showed a silica oxygen bond (Si-O-Si) at 810.1 cm^{-1} and a Si-OH bond of 3000-3500 cm^{-1} (Bassyoumi and Saleh, 2023). The strong indication of silica content and its functional groups symbolized by the FTIR result confirmed the fulfillment of the research objective related to the effectiveness of the sol-gel method in producing nano-silica.

3.5 SEM Test Results

Figure 5 shows the fly ash and nano-silica image with 10000x magnification taken by SEM. The porosity of fly ash is identified in Figure 5a. Meanwhile, in the 5b section, there is an image of nano-silica morphological structure in the form of aggregates or agglomerates. The morphology of the particles indicates a tendency for silica samples to aggregate, suggesting that additional dispersion may be required for applications requiring isolated particles (Figure 5b). Finally, the SEM images implied that silica nanoparticles' aging time and size are

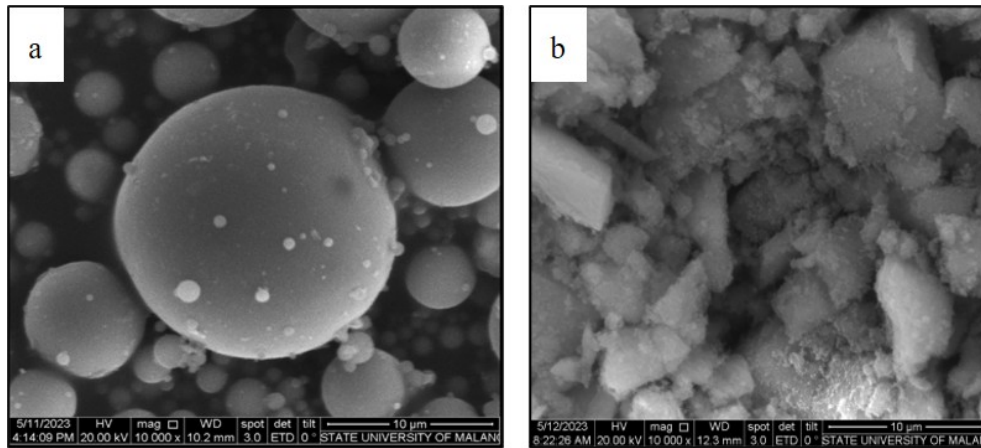


Figure 5. SEM Test Results (a) Fly Ash Magnification 10000 \times ; (b) Nano-Silica Magnification 10000 \times

essential to product morphology.

4. CONCLUSIONS

Based on the results of multiple tests and analyses, it can be concluded that the selected operating conditions, including solution molarity, the solution-to-fly ash ratio, and the silica synthesis method, significantly impacted the purity of the produced nano-silica. This research found that the selected process could produce promising silica products with high purity levels within the range of 97.47% to 98.57%. In addition, silica nanoparticles have great particle sizes ranging from 151.2 nm to 295.4 nm, along with smooth morphology and relatively well-distributed particles.

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