

Thermodynamic Modeling and Experimental Studies of Bayerite Precipitation from Aluminate Solution: Temperature and pH Effect

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ABSTRACT: Bayerite is one of the phases of aluminum hydroxide which is precipitated by the carbonation of aluminate solutions obtained from sintered nepheline syenite leaching. In this study, the conditions for the bayerite formation were predicted by thermodynamic modeling of the carbonation process and the Bromley- Zemaitis model was used for this purpose. Carbonation experiments were carried out at pH 11 and the temperature range of 50- 90 °C based on the data obtained from thermodynamic modeling results. XRD analysis of products showed that bayerite was the predominant phase at all temperatures. SEM and LDS analysis indicated that the bayerite precipitates had uniform morphology and bimodal particle size distribution with mean particle size of 4.6 μm at 50 °C to 12.9 μm at 90 °C. It was found that the d_{50} increased slowly at the precipitation temperature ranging from 80 to 90 °C, from 12.6 to 12.9 μm and the effect of temperature was on the shape of particles. XRF analysis of the products indicated that the amount of Al_2O_3 and SiO_2 in the bayerites decrease by increasing the temperature. According to the thermodynamic modeling data and experimental results, the temperature of 80 °C and pH 11 were determined as optimal conditions for bayerite precipitation.

KEYWORDS: Aluminate solution; Carbonation; Bromley-Zemaitis model; Bayerite.

INTRODUCTION

The lack of high grade bauxites in some countries drew attention to alternative alumina raw materials such as nepheline syenite, alunite and clay minerals. Nepheline syenite processing to produce aluminum hydroxide is similar to lime-soda sintering for the low-grade bauxite

ores processing. Unlike lime-soda sintering, large amounts of alkali will be produced with nepheline syenite[1, 2]. Gibbsite, bayerite, and nordstrandite are three important polymorphs of aluminum hydroxide [3]. The pH of the mother liquor controls the structure

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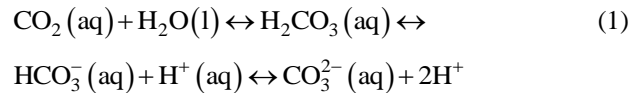
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of the aluminum hydroxide precipitate. Auto precipitation or injecting preheated carbon dioxide into an aluminate solution in acidic pH leads to the formation of gibbsite. Bayerite is formed with more rapid precipitation from cool or less alkaline pH. Neutral environments favor the formation of nordstrandite [4]. The formation of gibbsite is favored at higher temperatures, while bayerite is formed predominantly at room temperature [5]. Experimental data has shown that bayerite is the more thermodynamically stable polymorph of aluminum hydroxide because of the highest density and symmetry among the polymorphs of aluminum hydroxide [6].

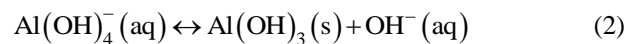
Mahin *et al.* (1913), Lajner (1961), Adamson (1963) and Rulnova (1968) have shown that the separation of aluminum hydroxide from aluminate solutions is based on the hydrolysis of the aluminate ion, $\text{Al}(\text{OH})_4^-$. This reaction reduces the pH of the solution due to the absorption of carbon dioxide which is usually called the carbonation process in alumina industry [7-9]. Czajkowski *et al.* (1981) discussed the effect of the CO_2 absorption rate on aluminate decomposition and developed a mathematical model of the carbonation process [9]. Zhou *et al.* (2009) investigated the agglomeration of gibbsite particles from carbonation process [10]. Li *et al.* (2009) proposed a crystallization process of aluminum hydroxide from the reaction of sodium aluminate and sodium bicarbonate solutions. All the aluminum hydroxides precipitated in the research were identified as gibbsite type [8]. Klimenko *et al.* (2013) comprehensively examined the effect of such as the pH value, the initial NaOH concentration, the temperature on the carbonation of aluminate solution and showed that $\text{Al}(\text{OH})_3$ always contains some dawsonite [11]. You *et al.* (2013) synthesized bayerite by the precipitation reaction of a sodium aluminate solution with sodium bicarbonate in a mixed suspension-mixed product removal crystallizer [12]. Li *et al.* (2013) produced aluminum hydroxide from liquor after desilication from nepheline syenite by injecting the mixture of air and carbon dioxide [1]. Yeboah *et al.* (2014) investigated a comparative study of the super cooling and carbonation processes of the bauxite and showed that alumina from carbonation process is larger than super cooling process [13].

When carbon dioxide is injected into an aluminate solution, first it reacts with water to produce carbonic

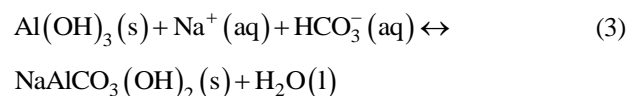
acid, which dissociates to H^+ and HCO_3^- ions. Then, HCO_3^- ion is dissociated to produce H^+ and CO_3^{2-} ions. Since carbon dioxide can twice dissociate in water, the reaction scheme may be written as [14-16]:



H^+ ions neutralize free OH^- ions in the aluminate solution. Since the reaction between H^+ and OH^- is instantaneous forming water, the reaction is very fast and free hydroxide is rapidly neutralized. Therefore, by decreasing the pH, the aluminate solution becomes unstable and aluminum hydroxide precipitates [2, 9]:



By injecting more gas, the amount of HCO_3^- ions increase and at pH values below 10 dawsonite ($\text{NaAlCO}_3(\text{OH})_2$) is formed and contaminates the valuable product [1, 11]:



The silica in the aluminate solution also contaminates the product. Rapid contamination of product with the silica is observed only at the end of the carbonation process [13]. In industrial conditions, carbonation process is carried out at temperatures from 70 to 80°C. At lower temperatures, fine-crystal aluminum hydroxide is produced [13].

In this study, thermodynamic modeling of aluminate solution carbonation was performed to precipitate aluminum hydroxide as a bayerite phase. The Bromley- Zemaitis model is used for calculation of activity coefficients which is combined in a commercial code OLI Analyzer Studio as the Aqueous model. Bromley (1973) replaced β coefficient of the Debye-Hückel model with a coefficient B ($B=B_+ + B_-$) to use the model for electrolytes with ionic strengths even greater than 6 mol/kg [17]. Zemaitis (1986), of OLI Systems Inc., added the C and D coefficients to the Bromley model, which is usable for electrolytes with ionic strength up to 30 mol/kg. The general model equation as follows:

Table 1: The parameters for calculating activity coefficients of aluminate solution.

B	C	D	I
$B_1 + B_2T + B_3T^2$	$C_1 + C_2T + C_3T^2$	$D_1 + D_2T + D_3T^2$	m_i, z_i

$$\log \gamma_{\mp} = -\frac{A|z_+z_-|\sqrt{I}}{1+\sqrt{I}} + \frac{(0.06+0.6B)|z_+z_-|I}{\left(1+\frac{1.5}{|z_+z_-|}I\right)^2} + BI + CI^2 + DI^3 \quad (4)$$

Where A , B , z_+ and z_- are the Debye-Hückel Constant, Bromley parameter, Charge of the cations and anions, respectively. C and D coefficients are Zemaitis parameters. I is the ionic strength which a function of the concentration of all ions present in the solution m_i and charge number of the ions z_i . The Bromley-Zemaitis equation is a function of temperature, ionic strength, and individual species concentrations [18]. The parameters for calculating activity coefficients of aluminate solution are presented in Table 1. In this study, no Debye-Hückel, Bromley and Zemaitis parameters were entered into the software other than those existing in OLI's default database. The OLI engine performs Gibbs Energy minimization to calculate the activity coefficients and determines, under equilibrium conditions, the most stable species in the solution, solid and gas phase. The Bromley-Zemaitis model is usable in the temperature range of -50 to 300 °C, pressure range of 0 to 1500 bar. The standard-state properties are calculated by fitting of the Helgeson-Kirkham-Flowers equation of state [19, 20].

However, a good literature review showed that there are a few publications on the carbonation process of aluminate solution obtained from sintered nepheline syenite leaching to produce aluminum hydroxide as a bayerite phase. Hence in this study, the authors modeled the thermodynamically aluminate solution at CO₂ concentration range of 0-50 g/l and temperature range of 50- 90 °C. Effect of carbon dioxide gas injection on solution pH as well as temperature effect on the formation of various products were investigated. At the following carbonation experiments were performed based on the data obtained from thermodynamic modeling results to produce only bayerite. To carry out experiments, actual aluminate solution obtained from alkaline leaching of sintered nepheline syenite was used.

Table 2: Chemical composition of aluminate solution.

Components	Concentration(g/l)
Al ₂ O ₃	10.5
K ₂ O	17.5
Na ₂ O	36.5
SiO ₂	0.12

EXPERIMENTAL SECTION

Aluminate Solution Preparation

The aluminate solution was obtained from alkaline leaching of sintered nepheline syenite (provided from Azarshahr alumina pilot in Eastern Azerbaijan province, Iran) at the temperature of 80 °C and atmospheric pressure in the laboratory. After desilication of aluminate solution at the temperature of 90 °C by milk of lime, carbonation process was carried out on the solution. The chemical composition of the aluminate solution is represented in Table 2.

Thermodynamic modeling

The aluminate solution was modeled by the Bromley-Zemaitis of OLI Analyzer software (Aqueous model) to predict the optimal conditions for bayerite precipitation. The effect of carbon dioxide gas concentration on solution pH as well as the effect of temperature on carbonation products in the concentration range of 0-50 g/L of carbon dioxide and temperature range of 50- 80 °C were investigated. For a determined set of conditions, the OLI engine performs Gibbs Energy minimization to determine the most stable species in the solution, solid and gas phase under equilibrium conditions.

Experimental procedure

Schematic diagram of experiment apparatus used the carbonation process is shown in Fig.1. Carbonation experiments were performed in 2.5 L glass reactor filled with 1.5 liters of solution. The solution was heated to the temperatures of 50, 60, 70, 80 and 90 °C. Carbon dioxide at a certain flow rate was injected through a bubbler ring with many holes into aluminate solution

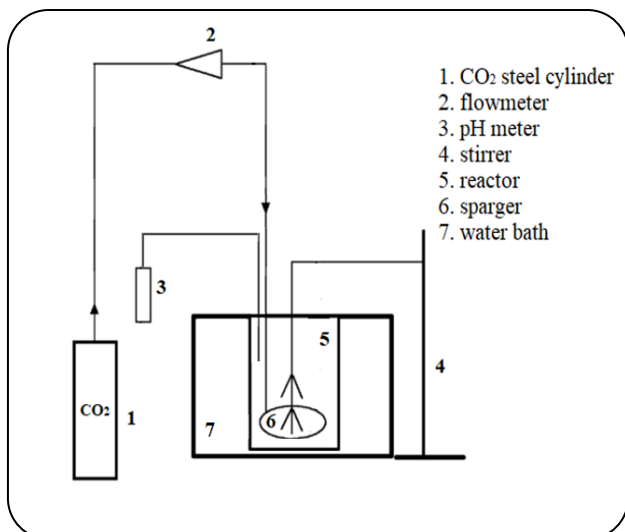


Fig. 1: Schematic diagram of experiment apparatus of carbonation process.

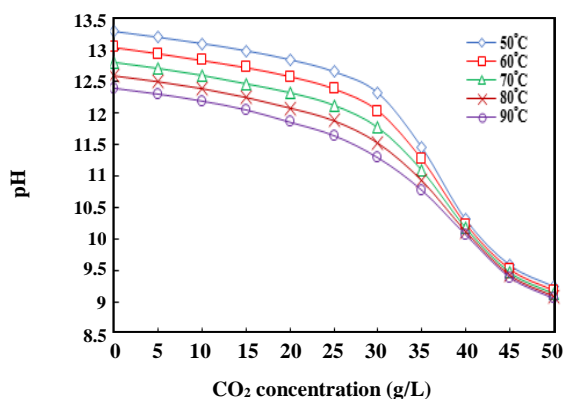


Fig. 2: The effect of CO₂ concentration on the pH of the aluminate solution.

and was stirred at 400 rpm. After reaching the desired pH, gas injection operation stopped. By cooling the solution, the possibility of crystallization of aluminum hydroxide provided. After 8 hours, carbonized solution was filtered and precipitate was washed with hot distilled water to remove the alkaline. The precipitates were dried in an oven at 50 °C for 48 hours. The precipitates were characterized using powder X-Ray Diffraction (XRD) to identify the major phases present, X-Ray Fluorescence (XRF) to determine the elemental composition, Scanning Electron Microscopy (SEM) to examine the morphology of the crystals, and the Laser Diffraction Spectrometry (LDS) to determine the distribution of particle size of products.

RESULTS AND DISCUSSION

Thermodynamic modeling results

The effect of CO₂ concentration on pH solution at different temperature

The one of the main factors determining the properties of the aluminum hydroxide is the pH of solution. In order to precipitation of bayerite by carbonation, at first, the effect of carbon dioxide of the pH solution at temperatures of 50, 60, 70, 80 and 90 °C investigated. A titration curve, pH versus CO₂ is shown in Fig. 2. It can be seen that the reducing pH has similar trend at all temperatures, but at 90 °C, due to the lower solubility of carbon dioxide in solution, the trend of reducing the pH is slower. Injecting CO₂ into the solution lowers the pH initially (according to reaction (1)). The pH does begin to slowly decrease after pH 10 because of the simultaneous dissociation of carbon dioxide gas in solution to generate H⁺ ions and OH⁻ ions production according to the reaction (2). Therefore, more carbon dioxide gas is needed to reduce pH.

The effect of pH and temperature on the stability of species in aluminate solution

Changes in the concentration of ions such as aluminate, hydroxide, bicarbonate and carbonate lead to the production of aluminum hydroxide and other carbonation products. The concentration variation of aluminate, hydroxide, carbonate and bicarbonate ions versus pH at the temperatures of 50-90 °C is illustrated in Fig. 3. At the beginning of the carbonation process, by injecting carbon dioxide gas, the amount of HCO₃⁻ ion in the solution increases and simultaneously neutralization of hydroxide ion occurs. Therefore, the amount of hydroxide ion in the solution reduces while concentration of aluminate ion remains constant. By further decrease of pH, at a pH about 11, amounts of OH⁻ and HCO₃⁻ ions become approximately equal. Hydrolysis of aluminate ions occurs in this range and aluminum hydroxide precipitates from the solution (reaction (2)) which is indicated by the change of the color of the solution from clear to opaque. After this pH, if carbon dioxide gas injection continues, the amount of CO₃²⁻ decreases whereas the concentration of HCO₃⁻ increases, therefore dawsonite according to reaction (3) forms which contaminates the aluminum hydroxide. Therefore, when hydrolysis of aluminate ions starts gas injection stops.

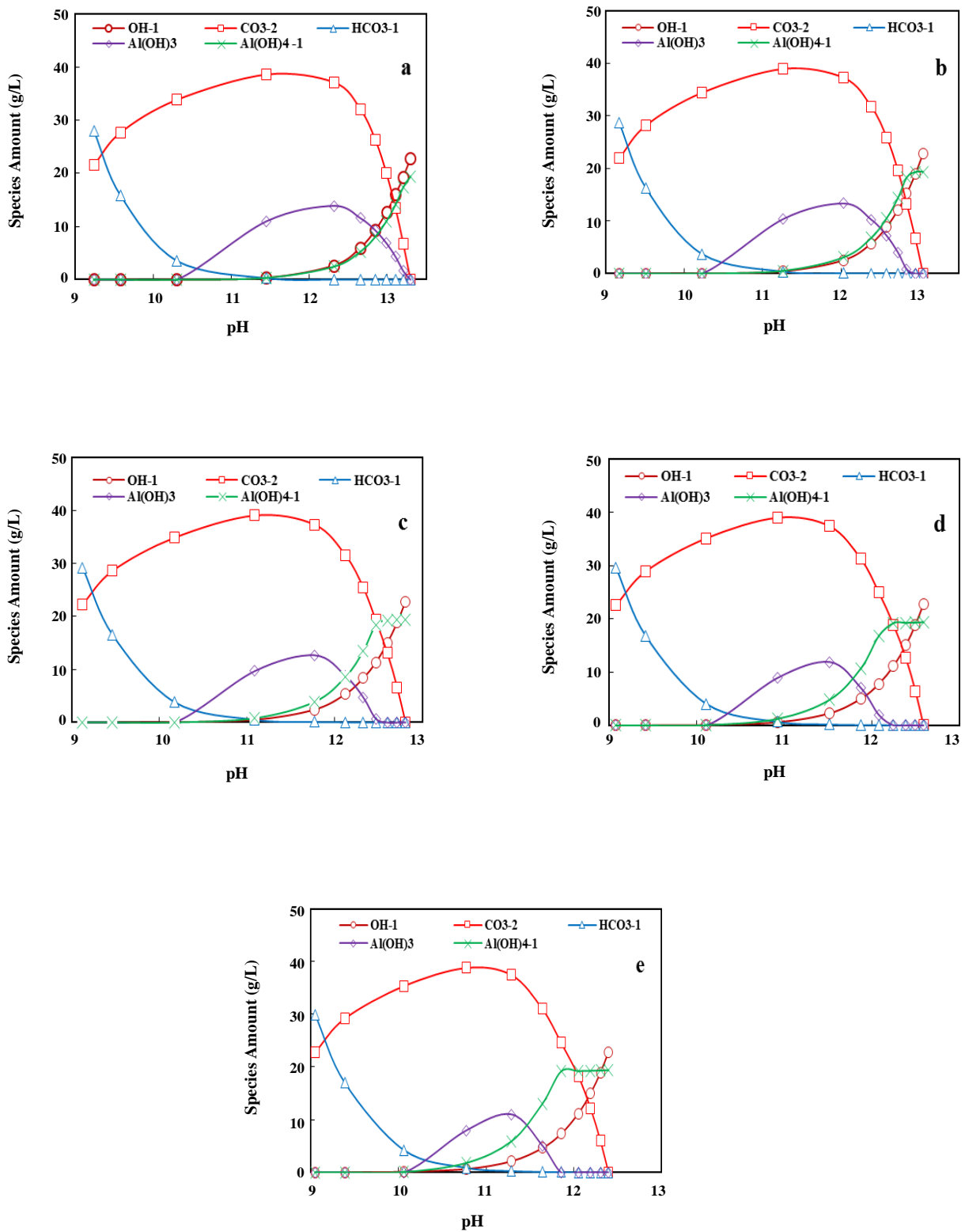
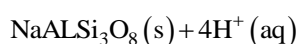
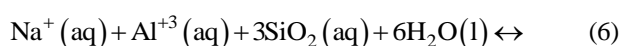
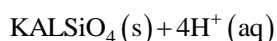
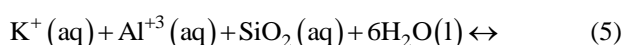


Fig. 3: Concentration variation of species in aluminate solution versus pH after carbonation; a)50 °C, b)60 °C, c)70 °C, d)80, e)90 °C.

These changes in the solution are the same for all temperatures, but the amount of species in the solution decreases with increasing temperature. At 50 °C, Reaction (1) is faster due to increased solubility of carbon dioxide in solution. Therefore, aluminum hydroxide precipitation occurs earlier than higher temperatures.

The effect of pH and temperature on the stability of solid phases

Carbonation products at pH values ranging from 9 to 13 and temperatures of 50-90°C is shown in Fig 4. As can be seen from this figure, in addition to the production of a valuable aluminum hydroxide phase as a bayerite, others invaluable phases such as dawsonite, panunzite, $KAlSiO_4$, or albitelow, $NaAlSi_3O_8$ are produced and contaminate the product:



At all temperatures, bayerite is a predominant phase of aluminum hydroxide and maximum amount of bayerite is precipitated at 70°C. By decreasing the pH to 10, the amount of dawsonite is increased and it is the predominant product. At 10 < pH < 11, a mixture of bayerite and dawsonite is formed. Panunzite precipitates with bayerite at pH and temperatures above 11 and 70 °C, respectively. At low pH and temperatures, albitelow is formed. At pH above 11, bayerite with the lowest amount of dawsonite and silica compounds can form. Amount of bayerite decreases at 80 and 90°C than other temperatures. The solubility of carbon dioxide in the solution is low, Therefore carbon dioxide decomposition reaction in the solution (Reaction (1)) and the reaction of aluminum hydroxide precipitation (Reaction (2)) are slow. Using the modeling results, it can be concluded that hydrolysis of aluminate ion and precipitation of aluminum hydroxide occurs at carbon dioxide concentrations about 35 gram per liter, pH above 11, and at all temperatures of 50- 80 °C.

Experimental Results

Based on thermodynamic modeling results, at pH above 11 and temperature range of 50-90 °C, bayerite

with the lowest amount of dawsonite and silica compounds can be formed. Using thermodynamic modeling results, Carbonation experiments performed out at pH 11 to produce pure aluminum hydroxide, bayerite phase. The products are characterized using analysis techniques such as XRD, LDS, SEM, and XRF.

The XRD analysis of the products is shown in Fig. 5. Bayerite is the major phase formed in the carbonation process of aluminate solutions at all temperatures. During the carbonation process (8 hours), the amount of hydroxide ion in the solution increases according to Reaction (2) which causes to increase the final pH of the solution 1.5 to 2 units (based on experimental observations). Therefore, if a small amount of dawsonite or silica compounds are formed during carbonation, they have dissolved again. Therefore, final products are only bayerite. By increasing temperature, the peaks are very sharp, indicating good crystallinity of the product at 80 °C.

Carbonation of aluminate solution directly influences the physical properties of the product, such as the mean particle size, the particle size distribution, and morphology. Particle agglomeration is the most important way to rapidly enlarge the particle size. The crystallization temperature of the solution plays the most important role in the agglomeration. Moreover, an increase in the crystallization temperature promotes agglomeration [9].

The particle size distributions of bayerite precipitates at 50, 60, 70, 80 and 90 °C are shown in Fig. 6. It is indicated that the bayerite precipitated from all experiments have the bimodal particle size and a mixture of fine and coarse particles. The particle size of bayerite increased with increasing temperature ranging from 50 to 90°C, the variation of mean particle sizes (d_{50}) as a function of precipitation temperature is shown in Fig. 7. It was found that the d_{50} increased rapidly at the precipitation temperature ranging from 50 to 80°C, from 4.6 μm to 12.6 μm , at the following, particle size increasing with temperature is slow. It is indicated from Fig.7 that with increasing temperature from 80 to 90°C the particle size of bayerite is increased from 12.6 to 12.9 μm . It means further increasing at a temperature above 80 °C to more improvement in particle size is not necessary. The percentage of particles of larger than 10 μm in bayerite precipitates are 8, 16, 48, 65 and 57 percent at temperatures of 50, 60, 70, 80 and 90 °C, respectively.

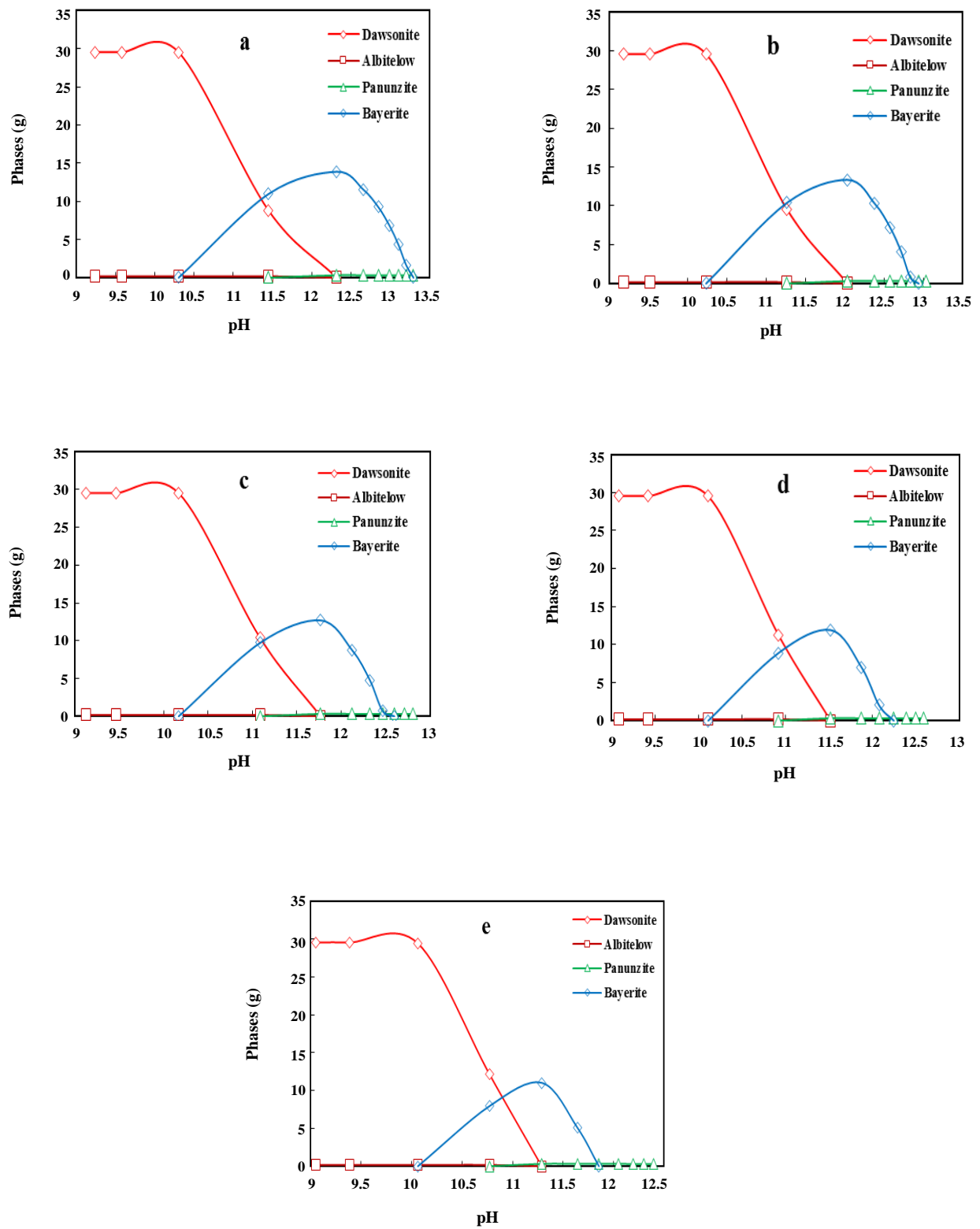


Fig. 4: Carbonation products versus pH at temperatures a) 50 °C, b) 60 °C, c) 70 °C, d) 80 °C, e) 90 °C.

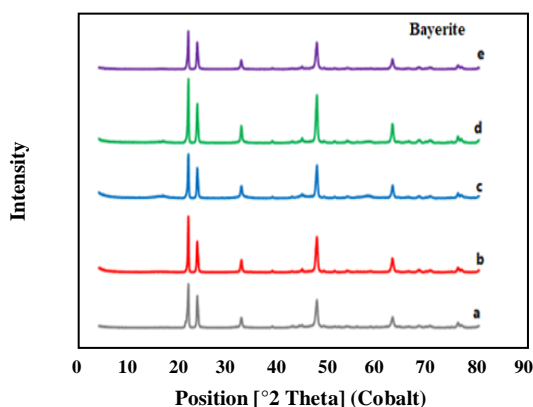


Fig. 5: XRD pattern of products of carbonation process at pH=11; a) 50 °C, b) 60 °C, c) 70 °C, d) 80, e) 90 °C.

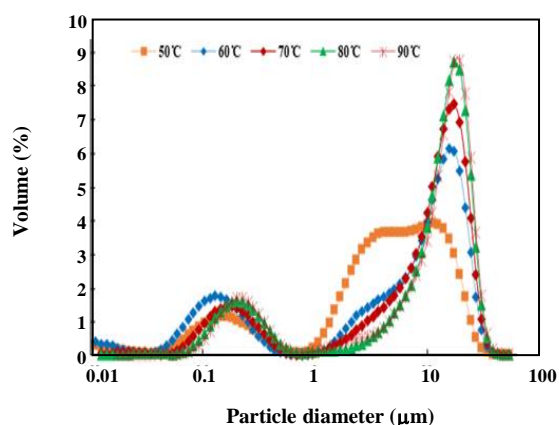


Fig. 6: Particle size distribution of bayerite precipitated.

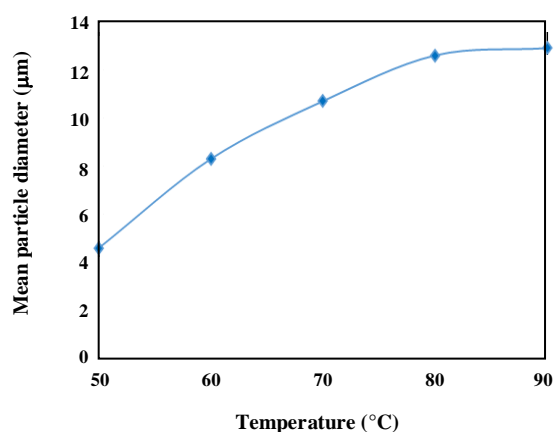


Fig. 7: Effect of temperature on the mean particle size (d_{50}) of bayerite precipitates.

Since the particle size of products is fine at temperatures of 50 °C and 60 °C, it is concluded that carbonation is carried out at a temperature above 60 °C. The SEM photographs of carbonation products at temperatures of 70, 80 and 90 °C are shown in Fig. 8. The bayerite precipitates have uniform morphology and consist of small spherical particles and large aggregates (ovoid-shaped). The small particles are agglomerated to form the large particles. The morphology of bayerite precipitates formed at 70 and 80 °C is the same, but at 80 °C agglomeration of particles is improved, hence the size of particles is increased. By increasing the temperature to 90 °C, in addition to small spherical and ovoid-shaped particles, bunches of ovoid-shaped particles are also seen. The SEM photographs indicate an effect of temperature on the shape of particles.

The final concentration of Al_2O_3 in spent solutions after carbonation for temperatures of 70, 80 and 90 °C is 4.7, 5.3 and 5.5 g/L and the yield of product is 55.2, 49.5 and 47.6 %, respectively. Chemical composition of carbonation products, bayerite, at 70, 80 and 90 °C is represented in Table 3. Results of XRF analysis of products showed that the amount of silica in the bayerite produced at 90 °C as well as the amount of Al_2O_3 is lower than the other temperatures. The amount of Al_2O_3 in products formed at 70 and 80 °C is about 67 %, but the amount of silica and other elements in bayerite obtained at 80 °C are less than 70 °C. According to the XRF analysis results, it can be concluded that the product has a higher purity at 80 °C.

CONCLUSIONS

The results of thermodynamic modeling of the carbonation process showed that the hydrolysis of aluminate ion and formation of aluminum hydroxide occurs at pH above 11 and bayerite is the predominant phase of aluminum hydroxide at temperatures of 50- 90 °C. By increasing the concentration of carbon dioxide, pH values lower than 10, bicarbonate ions increase in the solution, therefore the product is only dawsonite. Based on the thermodynamic modeling data carbonation experiments were carried out at pH 11 and temperatures of 50, 60, 70, 80 and 90 °C. Phase analysis study indicates that the major phase of all products is bayerite. Bayerite precipitates have uniform morphology, bimodal size and consisted of fine spherical particles and large agglomerate particles. Particle size

Table 3: XRF analysis of amounts of products produced from carbonation experiments.

Compounds (%)	Al ₂ O ₃	SiO ₂	CaO	Na ₂ O	K ₂ O	Fe ₂ O ₃	MgO	SO ₃	P ₂ O ₅	ZnO	Cl	L.O.I
A- 70°C	67.35	0.69	0.11	<0.1	0.15	-	<0.1	<0.1	<0.1	<0.1	<0.1	31.81
B- 80°C	66.58	0.58	0.12	<0.1	<0.1	-	<0.1	<0.1	<0.1	<0.1	<0.1	32.72
C- 90°C	63.90	0.25	0.14	0.30	0.4	0.04	0.01	0.04	-	0.004	-	32.25

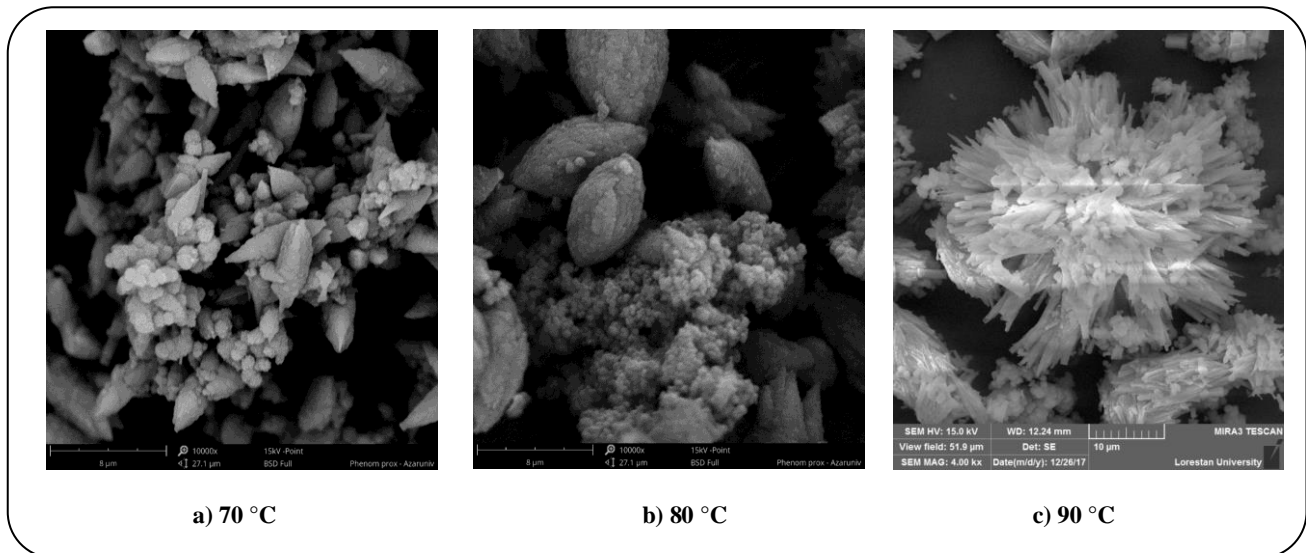


Fig. 8: SEM photographs of the bayerite precipitates.

distribution of the bayerite is bimodal at the temperature range of 50-90 °C. The mean particle size of bayerite precipitates increases from 4.6 to 12.9 μm by increasing temperature from 50 to 90 °C. The particle size at 90 °C is slightly larger than 80 °C. It is indicated that the effect of temperature increasing above 80 °C is on the shape of particles. The large particles are agglomerated and bunches of ovoid-shaped particles are formed. By increasing the temperature, the amount of Al₂O₃ and SiO₂ in the bayerites decreases. Generally, the product formed at 80 °C has a higher purity. According to thermodynamic modeling and experimental results, it is concluded that pH 11 and the temperature of 80 °C are suitable for bayerite precipitation.

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REFERENCES

- [1] Li Y.J., Lei T., Yang D.J., [Radionuclide of Process of Carbon Decomposition and Anneal of Liquor after Desilication from Nepheline](#), *App. Mech. Mater.*, (2013).
- [2] Panov A., Vinogradov S., Engalychev S., [Evolutional Development of Alkaline Aluminosilicates Processing Technology](#), *Light. Met.*, 9-16 (2017).
- [3] Karamalidis A.K., Dzombak D.A., ["Surface Complexation Modeling: Gibbsite"](#), John Wiley & Sons(JWS), (2011).
- [4] Schoen R., Roberson C.E., [Structures of Aluminum Hydroxide and Geochemical Implications](#), *Am. Mineral.*, 55(1970).
- [5] Lee M-y., Parkinson G.M., Smith P.G., Lincoln F.J., Reyhani M.M., [Characterization of Aluminum Trihydroxide Crystals Precipitated from Caustic Solutions](#), *J. Am. Chem. Soc. (ACS)*, (1997).
- [6] Wefers K., Misra C., [Oxides and Hydroxides of Aluminum](#), *Alcoa Technical Paper No. 19*, Alcoa Laboratories, (1987).

- [7] Misra C., "Industrial Alumina Chemicals", Am. Chem. Soc.(AC S), (1986).
- [8] Li Y., Zhang Y., Yang C., Zhang Y., Precipitating Sandy Aluminium Hydroxide from Sodium Aluminate Solution by the Neutralization of Sodium Bicarbonate, *Hydrometallurgy*, **98**(1): 52-57 (2009).
- [9] Czajkowski A., Noworyta A., Krótki M., Studies and Modelling of the Process of Decomposition of Aluminate Solutions by Carbonation, *Hydrometallurgy*, **7**(3): 253-261 (1981).
- [10] Zhou Q., Peng D., Peng Z., Liu G., Li X., Agglomeration of Gibbsite Particles from Carbonation Process of Sodium Aluminate Solution, *Hydrometallurgy*, **99**(3): 163-169 (2009).
- [11] Klimenko A.A., Shapovalov V.V., Kolesnik T.V., Shapovalova T.V., Osovska A.A., The Question of the Mechanism of Allocation Aluminum Hydroxide from Solutions of Sodium Aluminate, *J. Sci. Donetsk. Inter. Tech. Uni.*, 14-21 (2013), in Russian.
- [12] You S., Li Y., Zhang Y., Yang C., Zhang Y., Synthesis of Uniformly Spherical Bayerite from a Sodium Aluminate Solution Reacted with Sodium Bicarbonate, *Ind. Eng. Chem. Res.*, **52**(36): 12710-12716 (2013).
- [13] Yeboah I., Addai E. K., Acquah F., Tulashie S. K., A Comparative Study of the Super Cooling and Carbonization Processes of the Gibbsitic Ghanaian Bauxite, *Int. J. Eng. Sci. Inno. Tech.*, (2014).
- [14] Hunter K.A., "Acid-base Chemistry of Aquatic Systems", Dunedin, New Zealand: University of Otago, (1998).
- [15] Carroll J.J., Mather A.E., The System Carbon Dioxide-Water and the Krichevsky-Kasarnovsky Equation, *J. Solution Chem.*, **21**(7): 607-621 (1992).
- [16] Pahlevanzadeh H., Mohseni Ahooei A., Estimation of UNIQUAC-NRF Model Parameters for NH₃-CO₂-H₂O System, *Iran. J. Chem. Chem. Eng. (IJCCE)*, **24**(1) 21-26 (2005).
- [17] Bromley L.A., Thermodynamic Properties of Strong Electrolytes in Aqueous Solutions, *AIChE Journal*, **19**(2) 313-320 (1973).
- [18] Linz D., Rafal M., Berthold J., "Introduction to OLI Electrolytes", OLI Systems Inc., 1-21(2003).
- [19] "A Guide to Using OLI Studio Including Studio Scale Chem Version 9.1", OLI Systems Inc., (2014).
- [20] Zemaitis J.F., Clark D.M., Rafal M., Scrivner N.C., "Handbook of Aqueous Electrolyte Thermodynamics: Theory & Application", John Wiley & Sons (JWS), (2010).