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Effect of impact – reflective super – speed grinding on TanRai Aluminum hydroxide characteristics and dissolution efficiency in acid and base media

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ABSTRACT

Study on different characteristics of TanRai aluminum hydroxide dissolution process in acid and base media were carried out on unactivated and activated samples using 3000rpm centrifugal grinding method under other conditions. The experimental results showed that the centrifugal grinding process did not change the solid phase structure (Gibbsite), and the specific surface of the sample was increased by 1.5 times, average particle size was increased by 10 times, the surface morphology during process of TanRai aluminum hydroxide dissolution were different. In the case of the process of $Al(OH)_3$ dissolution in NaOH media, the corrosion reaction occurs on the all surface of the particle. It goes deep and accesses into the particle. The specific surface might be increased by 10 times in reaction processing, so the grinding did not increase recovery efficiency. In case of dissolution $Al(OH)_3$ in HCl media, the reaction occurs on surface particle so the grinding effects to dissolution recovery that increasing from 2 to 5 times.

Introduction

Currently, in the TanRai plant Bayer technology is used for producing Alumina (Al_2O_3). In particular, the stage of converting $Al(OH)_3$ into Al_2O_3 products requires a large amount of thermal [1]. Therefore, the cost of finally TanRai plant alumina is high and it is difficult to compete with products of the same type of Chinese companies. However, the processing of the intermediate product, aluminum hydroxide, is still profitable, so Vietnam National Coal-Mineral Industries Holding Corporation Limited-Vinacomin encourages the conversion of the intermediate product $Al(OH)_3$ into commercial material to improve economic efficiency. Therefore, it is the requirement that need for processing $Al(OH)_3$ into a widely used commercial product, thereby increasing economic efficiency and improving competitiveness. Now, there are 2 plants producing Poly Aluminum Chloride (PAC) coagulation on an industrial scale in Vietnam, are: the 10% Al_2O_3 liquid PAC production factory with a capacity of 25 thousand tons/year, is a branch of Joint Stock South basic chemicals; the second is a PAC factory with a capacity of 45 thousand tons/year, producing two types of liquid PAC with 17% Al_2O_3 and PAC powder with 30% Al_2O_3 under Viet Tri Chemical Joint Stock Company. These factories have used pure domestic materials $Al(OH)_3$ to produce PAC.

There have been many published works related to research on the dissolution of aluminum hydroxide in different conditions [2-8]. As is known, due to the duality of aluminum hydroxide, which is capable of dissolving in both acidic and basic environments

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[2,3,8], the mechanism and parameters affecting the dissolution process are also different.

The relationship between solid phase characteristics (surface morphology, surface area, particle size, structural bonding, etc.), dissolving mechanism, and reaction rate has been studied in the past [9], although it is far from complete.

The characteristics of the morphological change of the solid phase surface in the dissolution process, as well as the influence of the specific surface, particle size on the dissolution efficiency, were not mentioned in earlier research. This relationship is one of the most significant factors to consider when selecting activation methods to improve the process' efficiency and rate.

Therefore, in this paper, have been studied the morphological characteristics of the surface in the dissolution process with hydrochloric acid, sodium hydroxide and the influence of the grinding process on the aluminum hydroxide properties and dissolution efficiency of the above processes.

Experimental

Materials

The content of $Al(OH)_3$ TanRai in the original material is 93. %, Fe_2O_3 is 0.7%, and humidity is 6.2%. The particle size contribution (107 μm) is more uniform, and the specific surface area is 0.089 m^2/g.

Methods

Grinding techniques [10]

The basic working principle of the machine is to use the impact process with extremely high kinetic energy to split the raw material particles, and at the same time some of this kinetic energy is converted into heat capacity to dry the product and to shift and change the crystal structure of the crushed material. By having the grinding rotor use a large rotational speed of n > 3000 rpm and the machine has 3 grinding discs installed in series, one grain of raw material will be crushed through 3 mills. With this principle, it is very effective to crush aluminum hydroxide, which is a relatively moist and not very solid material with a layered structure, the material entering the mill does not need to be preheated. due to impact with great kinetic energy, aluminum hydroxide particles will not only separate in layers but also break in many different directions, will not only split in layers, but also break in different dimensions. Due to the high crushing efficiency, the period of samples staying in the grinding chamber is quite short so as to avoid changes in sample nature.

In this report, the ultra speed hammer centrifugal grinder at KimNguu Instrument & Chemical EM_IM.JSC (KIMEX JSC) was used. $Al(OH)_3$. The $Al(OH)_3$ Tanrai sample was ground at a speed of 3000 rpm. After grinding, the sample was dried at $100^oC \pm 5^oC$ for 2 hours and determined composition phase, particle size, the specific surface area.

Determining a material's characteristic structure and composition

- Measurement of surface area by Brunauer, Emett and Teller (BET);
-Measurement of particle size distribution by Laser scattering method;
-Siemens D5000 X-ray Powder Diffraction;
-Determination of aluminum content by complexon titration.

Procedure of $Al(OH)_3$ TanRai dissolution in HCl and NaOH solutions

Chemicals and intrumentals : HCl 36-38 % (PA, d=1,18 g/cm^3), NaOH 99,95% (PA), EDTA standard solution 0.05N, xylenol orange indicator , $ZnCl_2$ solution 0,05N, Electromagnetic balance with accuracy $\pm 10^{-4}$ g; heated bath, magnetic stirrer heater, mechanical stirrer… Experimental: The dissolution investigating of the sample before and after grinding was carried out in a solution of hydrochloric acid with a concentration of 3M and in a solution of NaOH with a concentration of 2M at 65°C, 75°C, 85°C and 95°C under the following conditions: Volume of aluminum hydroxide: 10 g, Volume of solution: 500 ml, Stirring speed: 250 rpm, Reaction time: 90 minutes.

The dissolving experiment is carried out in a glass bottle submerged in water. The sample is filtered once the reaction time has passed. Part of the filtrate is analyzed for the concentration of aluminum (Al^{3+}). The solid residue was washed, dried to determine the surface area obtained by scanning electron microscopy and to measure the particle size distribution.

Results and discussion

The modification of physical properties of grinded sample

After grinding, the physical properties of the TanRai $Al(OH)_3$ sample were changed. The results of particle size and surface area of $Al(OH)_3$ powder are shown in Table 1.

Table 1: Contribution of particle size and surface area results

Sample	Average particle size (μm)	Surface area (m^2/g)
Initial sample	107	0.089
Grinded sample	12	1.232

After the grinding process, the specific surface area was increased by 13.84 times for centrifugal grinding compared to the original sample. Because the particle sample has a layer structure, and is linked together by a weak hydrogen bond, the particle is broken with the layer separating mechanism, making a thin and flat shape. In the case of centrifugal hammer grinding, with the crushing mechanism, the particles are broken in different directions, resulting in a more rounded shape. This can be seen more clearly in the change of particle size.

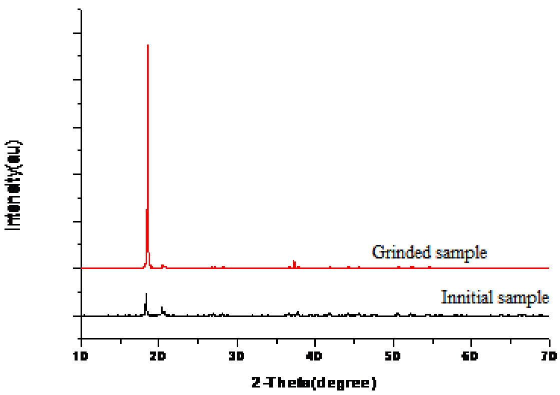


Figure 1: X-ray diffraction

By taking the samples' X-Ray diffraction (Figure 1) and comparing its result with the standard diffraction, the result showed that the structure of the initial and grinded sample has not changed. Both of them are Gibbsite structure [11].

Dissolution process of initial sample

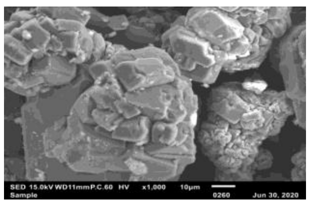
Table 2 shows the results of the TanRai $Al(OH)_3$ initial sample dissolution process in HCl and NaOH solutions. The data in Tab.2 shows that, after the reaction, the sample surface area tends to increase. However, the increase occurred similarly: with the same dissolubility rate (dissolubility efficiency) 27.01% in HCl and 28.83% in NaOH, the surface area increased from 0.089 m^2/g to 0.686 m^2/g in HCl and when dissolved in NaOH, the surface area of the sample increased dramatically from 0.089 m^2/g to 10.156 m^2/g.

Table 2: Dissolution of TanRai $Al(OH)_3$ initial sample in HCl and NaOH solutions

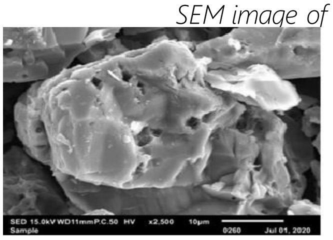
Solution	Reaction condition	H (%)	Surfae area (m^2/g)
Initial sample			0.089
HCl	3M, 85°C	27.01	0.686
NaOH	2M, 65°C	28.83	10.156
HCl	5M, 95°C	50.69	0.524
NaOH	2M, 75°C	48.77	5.701

A strong tendency to increase the sample surface area after dissolving in NaOH was also observed when comparing samples with a higher solubility rate. The sample surface area grew by roughly 6 times in HCl (from 0.089 m^2/g to 0.524 m^2/g) and 64 times in NaOH (from 0.089 m^2/g to 5.701 m^2/g) when the dissolution rate was 50.69 % with HCl and 48.77 % with NaOH. Furthermore, when the dissolution efficiency of both types of solutions increases to a certain extent, a tendency to reduce the surface area of the sample is observed. The surface area of samples in HCl decreased slightly from 0.686 m^2/g to 0.524 m^2/g. In the NaOH solution, the reduction was more clearly observed from 10.156 m^2/g to 5.701 m^2/g. This can be explained by the smoothness of the sample surface with greater dissolution, which leads to a significant decrease in the specific surface of the sample.

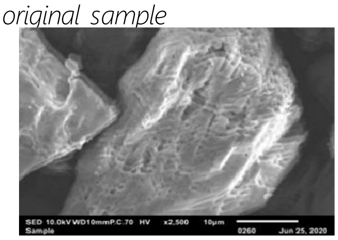
The tendency to drastically change the specific surface area of the sample in NaOH compared to that in HCl can be explained using SEM imaging (Figure 2).



SEM image of original sample



3M HCl, at 85°C, 27.01% effciency



2M NaOH, at 65°C, 28.83% effciency

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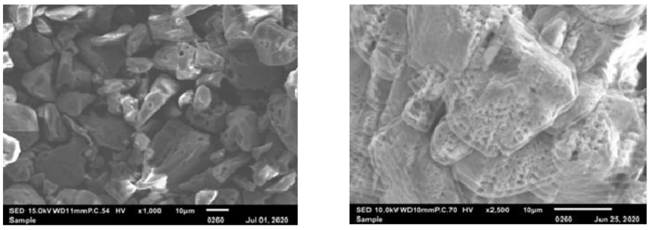

5M HCl, at 95°C, 50.69% effciency   2M NaOH, at 75°C, 48.77% effciency

Figure 2: SEM images of different dissolution conditions

The main phase composition in the sample is Gibbsite. Layer structures in Gibbsite are connected together by weak hydrogen bonds, producing in overlapping layers. The process of dissolving the sample in HCl occurs on the surface and has a deep cavity in some defects. However, the reaction of $Al(OH)_3$ with NaOH occurs by a different mechanism: The reaction occurs on the entire surface of the substance and tends to go deep evenly into the sample. On the surface of the solid phase has formed "holes, holes" with different sizes and depths.

Thus, with the initial (unrefined) sample, the surface area development during dissolution in NaOH occurs much more strongly than in HCl, but the reaction rate increases with increasing specific surface area. In other words, for the reaction in HCl, the surface area of the sample affects the dissolution efficiency more strongly than in NaOH. Therefore, in order to increase the dissolution efficiency of $Al(OH)_3$ in HCl, it is necessary to grind the sample before the reaction. The grinding process in addition to increasing the specific surface also gives the particles a "residual" energy, the particles in the solid phase will be in an excited state, thereby helping the reaction happen faster (higher efficiency).

Dissolution process of grinded sample in HCl solution

Table 3 : Results of aluminum hydroxide dissolution process in 3M HCl

Temp. (°C)	Initial sample		Grinded sample	
	Eff. (%)	Surface area (m^2/g)	Eff. (%)	Surface area (m^2/g)
Initial	0	0.089	0	1.232
65	6.73	-	34.61	0.875
75	14.12	0.325	45.78	0.951
85	27.01	0.686	65.07	-
95	33.99	-	73.64	-

The results of dissolution of grinded sample in HCl solution are shown in Table 3.

The results show that dissolution efficiency increased with temperature, dissolution efficiency with grinded sample being 2 to 5 times that of the initial sample at the same temperature. That is the proof of the effective use of the grinding process in the disolution of aluminum hydroxide in acids.

Dissolution process of grinded sample in NaOH solution

In order to confirm that the grinding process had little effect on increasing dissolution efficiency in NaOH, the aluminum hydroxide sample before and after grinding was dissolved under several different conditions. The obtained data are presented in Table 4.

Table 4: The results of TanRai aluminum hydroxide in 2M NaOH solution

Temp. (°C)	Initial sample		Grinded sample	
	Eff. (%)	Surface area (m^2/g)	Eff. (%)	Surface area (m^2/g)
Initial	0	0.089	0	1.232
65	28.83	10.156	33.35	1.047
75	48.77	5.701	47.91	0.524
85	82.61		78.17	
95	98.18		91.22	

The sample's before and after grinding dissolution efficiency was the same. As a result, the grinding procedure has no effect on $Al(OH)_3$ solubility in NaOH solution.

Conclusion

Conducted investigation and evaluation of the difference characteristics between the dissolution of aluminum hydroxide in NaOH and HCl, between the un-grinded sample and the grinded sample. The results showed that milling aluminum hydroxide in a centrifugal grinding did not change the phase structure, that the specific surface increased by 1.5 times, and that the average particle size decreased by 10 times. At the same time, the change of surface morphology during the dissolution of aluminum hydroxide in alkali and acid is different, leading to the effect of grinding on dissolution efficiency will be different too. In the case of dissolution by NaOH, the corrosive reaction takes place all over the surface of the particles, spreads deep into them, the specific surface can increase to more than 10 times during the reaction, so the grinding does not increase dissolution efficiency. In the case of dissolution with HCl, the reaction takes place mainly on the surface of the grain, so the grinding has a significant effect. The dissolution efficiency increases from 2 to 5 times, due to the increase of the specific surface of the solid phase.

The results show that the use of the centrifugal grinding machine before dissolving $Al(OH)_3$ TanRai will get better dissolution efficiency in PAC production in Vietnam

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