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Determination of the Optimum Conditions for the Production of Gamma Alumina (γ-Al_2O_3) By the Precipitation Method of the Sodium Aluminate Solution

Alaa D. Jawad Al-Bayati [*], Habaib A. Majeed Al-Taee [*] and Ibtehaj Faisal Abdulraheem [**]

[*] *Department of Chemical Industries, Institute of Technology Baghdad, Middle Technology University, Iraq*

[**] *Department of Chemical engineering, College of Engineering, University of Baghdad, Iraq*

Abstract

Design of experiments (DOE) was made by Minitab software for the study of three factors used in the precipitation process of the Sodium Aluminate solution prepared from digestion of α-Al_2O_3 to determine the optimum conditions to a produce Boehmite which is used in production of γ-Al_2O_3 during drying and calcination processes, the factors are; the temperature of the sodium aluminate solution, concentration of HCl acid added for the precipitation and the pH of the solution at which the precipitation was ended. The design of the experiments leads to 18 experiments.

The results show that the optimum conditions for the precipitation of the sodium aluminate solution which leads to the production of gamma alumina are 6.5 pH, Temperature of solution is 90 C and the concentration of the hydrochloric acid (HCl) is 21%. While all other conditions lead mainly to other phases of alumina which is mainly epsilon- alumina (ε-Al_2O_3).

Key Words: Gamma alumina, Precipitation, XRD

Introduction

Boehmite is the starting material for the production of gamma alumina by precipitation method [1]. Precipitation is one of the important steps used in the production of alumina by Bayer process. In the precipitation process the kinetics of the precipitation is very slow which need huge tanks for precipitation the liquor of diluted alumina. Large flow rate means the need of large residence time in the precipitation tanks i.e. large volume tanks [2, 3]. Precipitation of boehmite ($Al_2O_3.H_2O$) instead of gibbsite ($Al_2O_3.3H_2O$) in the process of alumina production is an energy saving [4]. Gamma alumina is one of the important materials used in industry as adsorbents, catalyst supports and catalysts in the form of Nano powder or as thin film coatings and in because it has large specific area, low cost, good thermal properties, surface acidity and interaction with deposed transition metals [5-11].

γ-Al_2O_3 is used in The production of boehmite by either the precipitation or hydrothermal methods needs to control the aluminum source, pH, temperature of the precipitation from the alumina liquor, aging, and the drying conditions and others affect the crystalline shape and size of the boehmite product [12]. The precipitation step in Bayer process needs 48-72 h and it is he limiting step in the production of alumina by Bayer process and precipitation method [13]. The method of precipitation, drying and calcination is one of the important methods of producing alumina from aluminium oxy-hydroxides [14]. All the stages used in the synthesis of aluminium hydroxides such as the solution pH, the concentration of the aluminium hydroxide in the liquor, temperature and many others has important effect on the properties of alumina produced [15]. Aluminium hydroxide is normally produced by the hydrolysis of the aluminium salts, alcoholates and from metallic aluminium. There are methods of precipitation the alumina from the salts by using ammonia treatment [16-18] and by precipitation with acid treatment [19].

Experimental Procedure

Sodium aluminate solution was prepared by digestion of 80 grams α-Al_2O_3 powder with one liter 3.5 M sodium hydroxide solution in 2 liter volume autoclave, the temperature for digestion is 190 C , absolute pressure 11.5 bar and the time for digestion is 3 hours. Design of experiments (DOE) was made by Mini Tab Statistical software to study 3 factors affecting the Precipitation of alumina from sodium aluminate solution by acid addition which is; pH at which ending the addition of the acid, temperature of precipitation solution and concentration of the HCL acid added.

The range of each factor is as shown in table (1). Table (2) shows the conditions of the 18 experiments obtained in the DOE. 50 ml of sodium aluminate solution is taken as a base for executing the acid precipitation for each experiment in the above DOE, the pH is tested by TS 625 pH meter, the temperature was regulated by magnetic stirrer heater and measured by mercury thermometer. The precipitate was filtered by 5 micron filter paper to separate the precipitate from the mother liquid and then washed several times with distilled water to remove all the contaminates and NaCL salt.

Table 1, Range of the factors studied

Serial	Factor	Minimum	Maximum
1	pH	6	9
2	Temperature °C	80	100
3	HCl%	5	37

Table 2, Conditions of the experiments of DOE

Run	Name of precipitate sample	pH	Temp. °C	HCL%
1	Pdf-1	7	80	21
2	Pdf-2	7	90	21
3	Pdf-3	8	84	11.5
4	Pdf-4	7	90	21
5	Pdf-5	6.5	96	11.5
6	Pdf-6	7	90	37
7	Pdf-7	8	96	11.5
8	Pdf-8	8	96	30.5
9	Pdf-9	6	84	30.5
10	Pdf-10	6	96	30.5
11	Pdf-11	7	90	21
12	Pdf-12	8	84	30.5
13	Pdf-13	6.5	90	21
14	Pdf-14	7	100	21
15	Pdf-15	7	90	5
16	Pdf-16	8	90	21
17	Pdf-17	7	90	21
18	Pdf-18	7	90	21

The precipitate was dried at 120 C for 4 hours in Memmert oven, and then calcined at 550 C in a furnace with a calcination program as shown in table (3).

The calcination program was chosen according to our experience and literature [20-21] XRD tests were made to all the calcined precipitates by XRD instrument using CuKα at 40 kV and 30 mA.

Table 3, Calcination program for alumina production from sodium alumina acid precipitation

Serial	Temperature ^{0}C	Time minutes
1	200	60
2	280	45
3	300	5
4	325	5
5	350	5
6	375	5
7	400	5
8	425	5
9	450	5
10	475	5
11	500	5
12	525	5
13	550	150
Total time for calcination	5 hr. +5 min.	

Table (4) show the maximum three peaks for each of the alumina and the phase obtained from the DOE we made.

Table (5) show the maximum three peaks of the alumina phases [22].

Table 4, the strongest 3 XRD peaks (2θ) and type of alumina phase obtained in each experiment of the DOE.

No.	Precipitate Name	2θ for the strongest 3 XRD peaks	Alumina phase
1	Pdf-1	45.937,46.51,36.95	Epsilon ε
2	Pdf-2	45.937,46.51,36.95	Epsilon ε
3	Pdf-3	45.937,46.51,36.95	Epsilon ε
4	Pdf-4	45.937,46.51,36.95	Epsilon ε
5	Pdf-5	45.937,46.51,36.95	Epsilon ε
6	Pdf-6	45.937,46.51,36.95	Epsilon ε
7	Pdf-7	45.937,46.51,36.95	Epsilon ε
8	Pdf-8	45.937,46.51,36.95	Epsilon ε
9	Pdf-9	45.937,46.51,36.95	Epsilon ε
10	Pdf-10	45.937,46.51,36.95	Epsilon ε
11	Pdf-11	45.937,46.51,36.95	Epsilon ε
12	Pdf-12	45.937,46.51,36.95	Epsilon ε
13	Pdf-13 Pdfg-13-D	66.76,23.44,37.68 48.87,28.03,38.40	ɤ-alumina boehmite
14	Pdf-14	45.937,46.51,36.95	Epsilon ε
15	Pdf-15	45.937,46.51,36.95	Epsilon ε
16	Pdf-16	45.937,46.51,36.95	Epsilon ε
17	Pdf-17	45.937,46.51,36.95	Epsilon ε
18	Pdf-18	45.937,46.51,36.95	Epsilon ε

Table 5, 2θ for the strongest 3 XRD peaks of some standard alumina phases [22]

Alumina phase	d-spacing for the strongest 3 XRD peaks	2θ
Epsilon ε	1.979,1.947,2.451	45.81,46.61,36.63
Gamma	1.4,1.98,2.39	66.76,45.78,37.60
Alpha	2.08,2.55,3.479	43.36,35.13,25.58

Results and Discussions

Figures 1-19 show the XRD peaks for the calcined samples obtained in our acid precipitation study of the sodium aluminate solution to produce ɤ-Al_2O_3. It is noticed that the conditions of 6.5 pH, 90 ^{0}C temperature and 21% HCl acid concentration leaded to production of boehmite after drying (4 hours @ 110 ^{0}C and 1 hr. @200 ^{0}C and 45 minutes @ 280 ^{0}C)

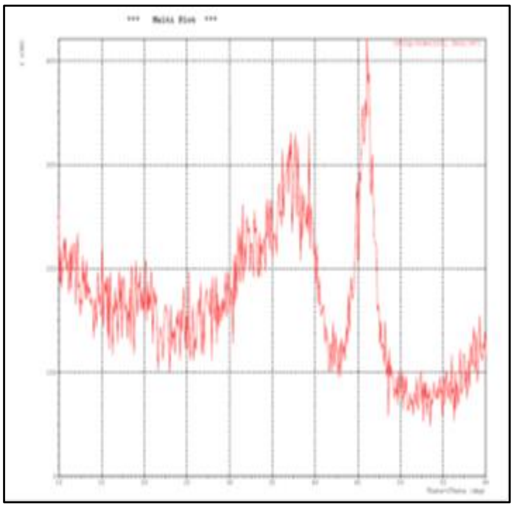

Fig. 1, XRD for Pdf-1

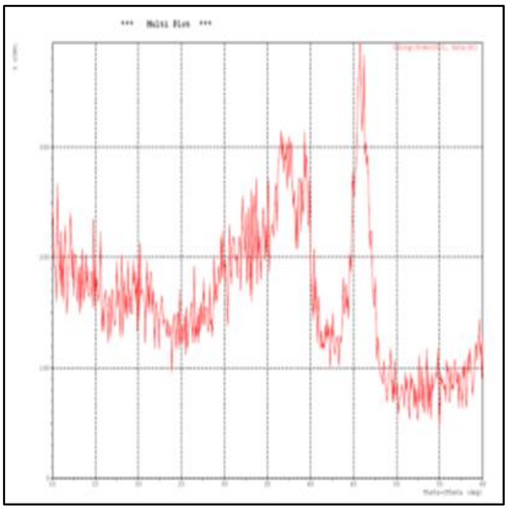

Fig. 2, XRD for Pdf-2

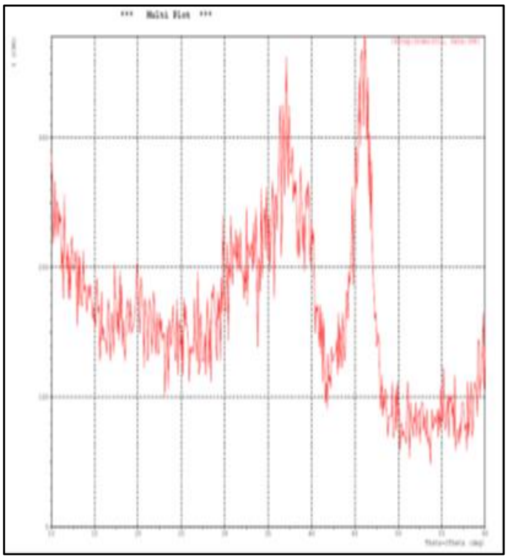

Fig. 3, XRD for Pdf-3

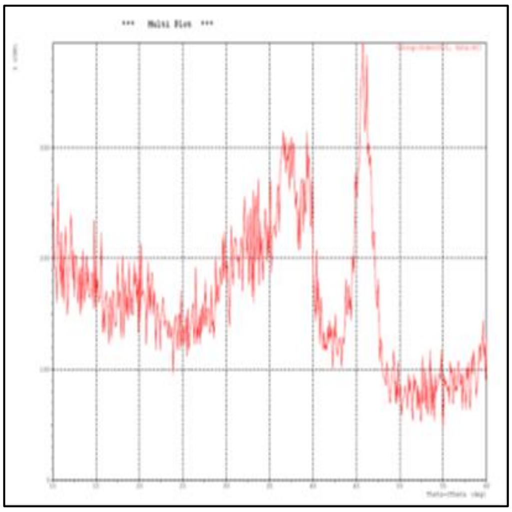

Fig. 4, XRD for Pdf-4

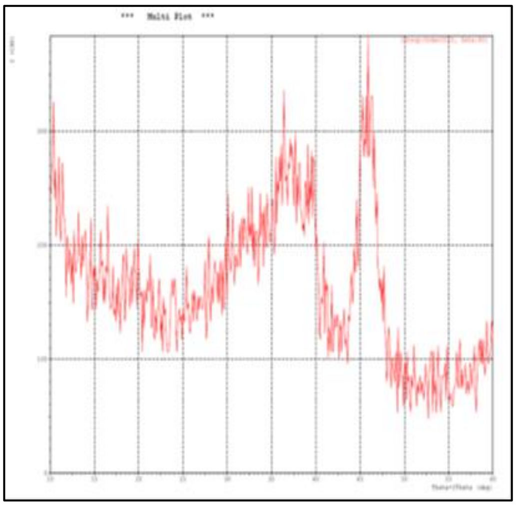

Fig. 5, XRD for Pdf-5

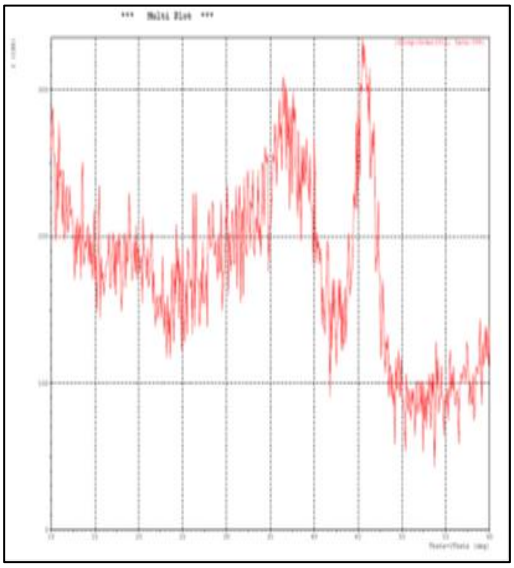

Fig. 6, XRD for Pdf-6

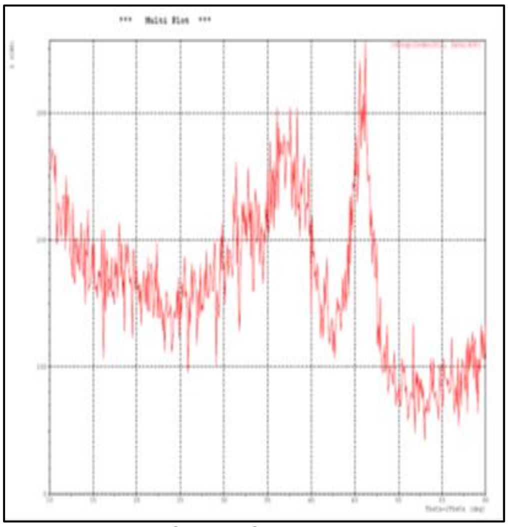

Fig. 7, XRD for Pdf-7

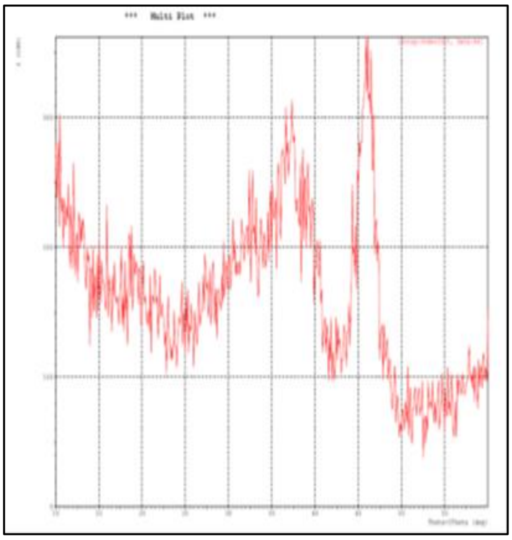

Fig. 8, XRD for Pdf-8

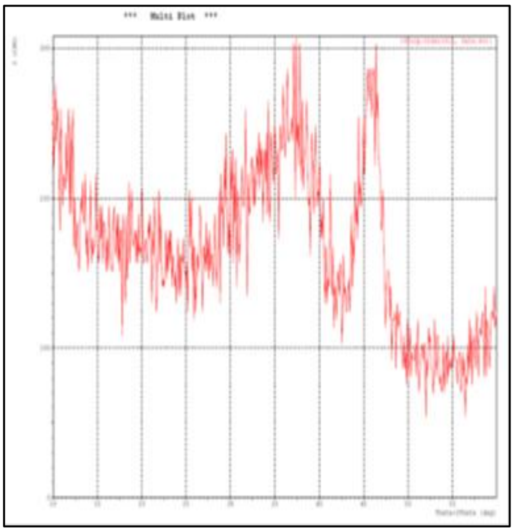

Fig. 9, XRD for Pdf-9

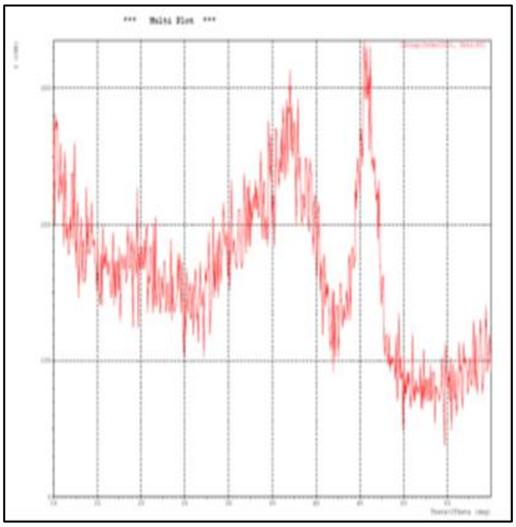

Fig. 10, XRD for Pdf-10

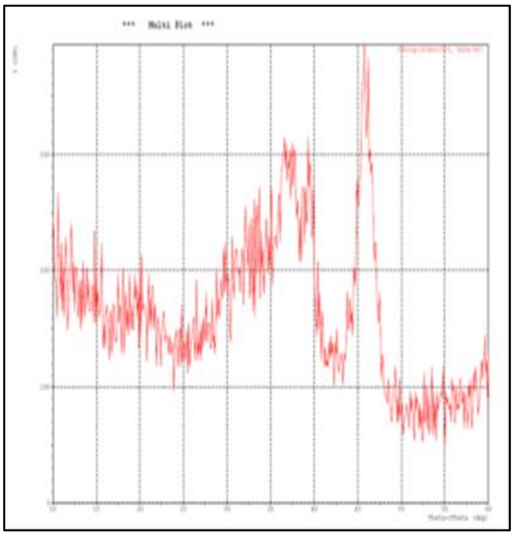

Fig. 11, XRD for Pdf-11

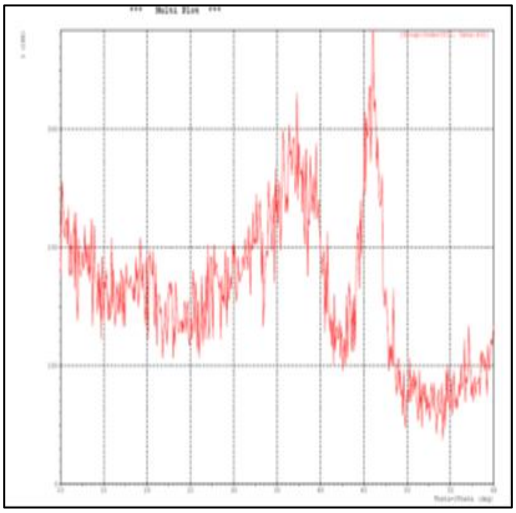

Fig. 12, XRD for Pdf-12

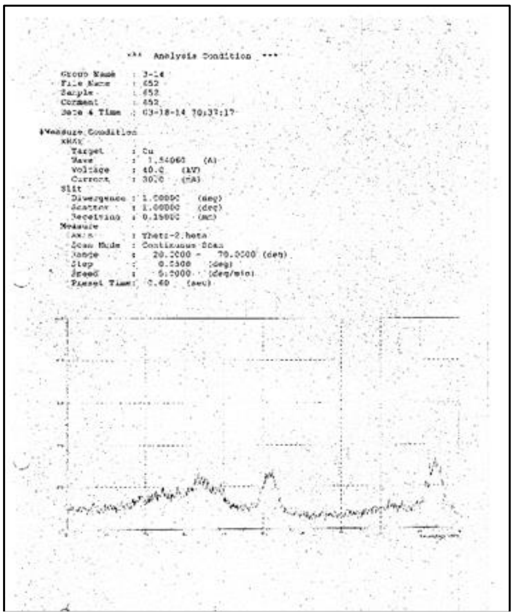

Fig. 13, XRD for Pdf-13

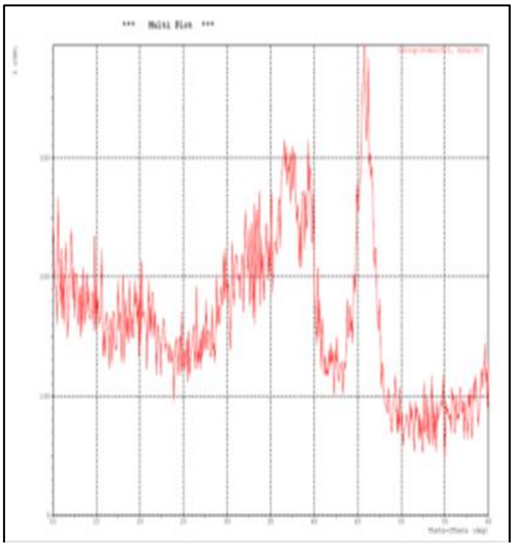

Fig. 14, XRD for Pdf-14

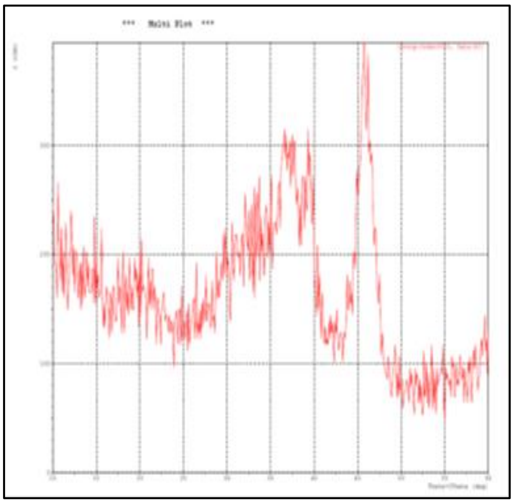

Fig. 15, XRD for Pdf-15

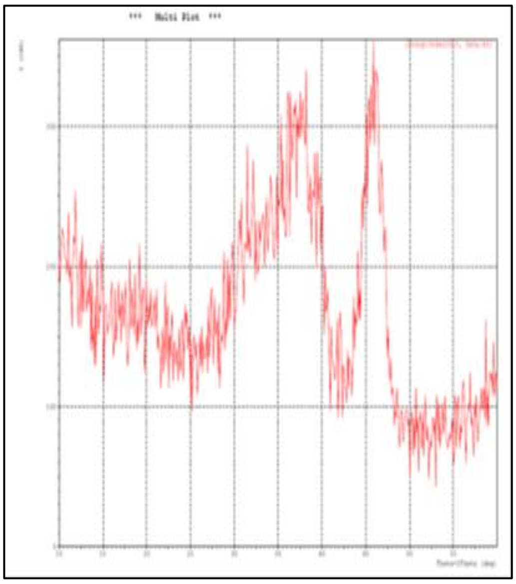

Fig. 16, XRD for Pdf-16

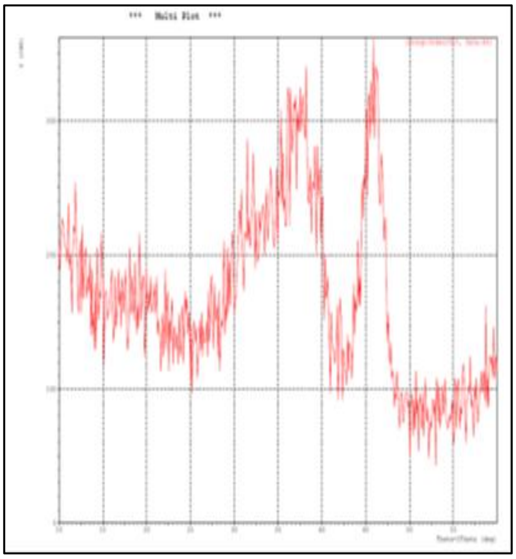

Fig. 17, XRD for Pdf-17

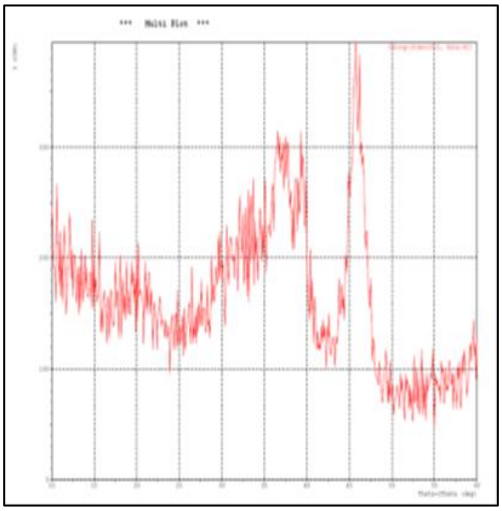

Fig. 18, XRD for Pdf-18

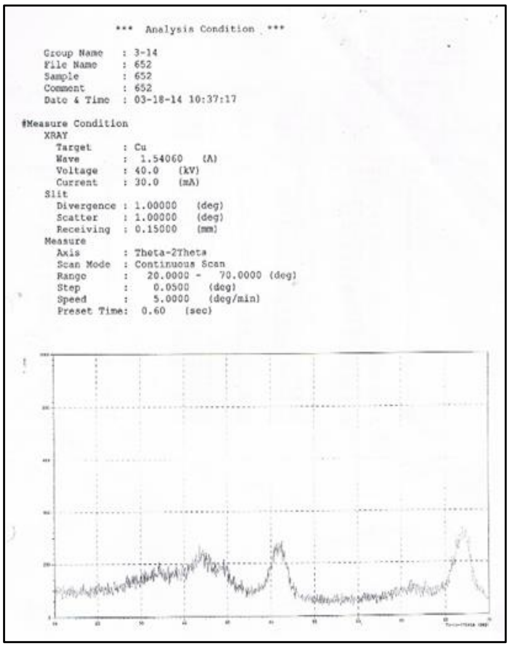

Fig. 19, XRD for Boehmite obtained from pdf-13 precipitation conditions and drying (4 hours @ 110 ^{0}C and 1 hr. @200 ^{0}C and 45 minutes @ 280 ^{0}C)

Conclusion

1- This work confirmed that the optimum conditions which lead to gamma alumina is very critical and need to be precise to precipitate boehmite and then to produce gamma alumina.

2- As shown in figure 13, the optimum conditions of pH 6.5, Temperature 90 ^{0}C and the HCL concentration is 21%. leads to Boehmite during precipitation

3- The digestion of α-Al_2O_3 with 3.5 M sodium hydroxide solution in autoclave at temperature of 190 ^{0}C, 11.5 bar absolute pressure and 3 hours the time for digestion was succeeded in conversion α-Al_2O_3 to ɤ-Al_2O_3 and it is compatible with the well-known literature of chemistry of Bayer process.

4- The DOE results shows that the interaction effects between the factors studied during precipitation is very important, and even any one or two factors are the same, the type of the alumina precipitated is different.

5- The use of α-Al_2O_3 as a source for producing ɤ-Al_2O_3 is promising in the production of boehmite and gamma alumina.

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References

1- R. Tipakontitikul, A. Niyompan, K. Srisurat, N. Kanchanarat, T. Tunkasiri, Journal of Micros copy Society of Thailand 22 , 20-22 (2008).

2- Barsha Dash, B.C.Tripathy, I.N.Bhattacharya and B.K.Mishra, The Royal Society of Chemistry, DaltonTrans., 39, 9108–9111(2010).

3- Parida, K.M, Pradhan, A. Maresh C., Das, J and Sahu Nrup,"Synthesis and characterization of nano-sized porous gamma alumina by controlled precipitation method", Materials Chemistry and Physics, 113(2009)244-248.

4- http://www.alteoalumina.com/en/calcined-aluminas.

5- Kotanigawa, T, Yamamoto, M.,Utiyama,M., Hattori, H. Tanabe, K., J. Applied Catalysis , ,1(7), 185-200 (1981).

6- Beguin B., Garbowski E., Primet M., J. of Catalysis, ,127(2), 595-604 (1991).

7- Peng X S. Zhang L.D, Meng G. W., J. Phys Chem B, 106(43), 11163-11167 (2006).

8- Lee H. C. ,Kim H. J. ,Chung S. H., J. Am Chem Soc, 125(10), 2882-2883 (2003).

9- K.A. De Friend, M.R. Wiesner, A.R. Baron, J. Membr Sci. vol. 224, 11-28 (2003).

10- Y. Cho, K. Han, K. Lee, ,J. Membr Sci. 104, 219 (1995).
11- X. Changrong, W. Feng, M. Zhaojing, L. Fanqing , P. Dingkun, M. Guangyao, J. Membr Sci. 116, 9-16 (1996).
12- Petrovice, R., Miynlonjic, S., Jokanovic, V., Kostic-Gvozdenovic, Lj., Petrovic-Prelevic, I., and Janackovic, Dj. , Powder Technology, **1**33, 185-189 (2003).
13- Chen, G.H., Chen, Q.Y., Yin, ZL. Zhang,B., Human Metallurgy ,31(1), 3-6 2003.
14- Bokhimi X., Sanchez-Valente J., Pedraza F., Journal of Solid State Chemistry, 166(1), 182-190 (2002).
15- V.A. Dzisko, A.S. Ivanova, and G.L. Vishnyakova, Kinetika I Kataliz,11, 483 (1970)
16- S. Music, D. Dragcevic, S. Popovic, and N. Vdovic, Mater. Chem. Phys., 59, 12 (1999)
17- V.K. Singh and R.K. Shina, Mater. Lett., 18, 201(1994)
18- N. Idrissi-Kandri, A. Ayral , Ch. Guizard, et al., Mater. Lett, **400**, 52 (1999)
19- G.L. Vishnyakova, V.A. Dzisko, L.M. Kefeli et al., Kinetika I KAtaliz**, 11,** 1541 (1970)
20- Al-Bayati, Alaa D. J, PhD thesis,"Manufacturing and description of platinum alumina used in reforming units of petroleum refineries' 2011, Syria. Al-Baath University.
21- Hlavac, J. ' The Technology of Glass and Ceramics an Introduction', 4th Edition, Elsevier Scientific Publishing Company, New York, 1983.
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