

The effect of precipitation pH and temperature of The Mg/Al Hydrotalcite synthesis on the glucose isomerization

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ABSTRACT

The application of glucose-isomerase as a biocatalyst in glucose isomerization requires a long time and complex conditions. Mg/Al hydrotalcite is considered to substitute the utilization of enzyme as the catalyst in this process. Mg/Al hydrotalcite was generated by coprecipitation of Mg(NO3)2.6H2O and Al(NO3)3.9H2O. The crystal quality of Mg/Al hydrotalcite is influenced by pH and temperature during the synthesis process. We evaluated the catalytic performance of Mg/Al hydrotalcite which is generated on various precipitation pH and temperatures by isomerizing glucose into fructose. The Mg/Al hydrotalcite was carried out in Mg/Al molar ratio 3:1 for 18 h. The characteristic of the Mg/Al hydrotalcite catalyst was done Fourier Transform Infra-Red (FTIR), X-Ray Diffractometer (XRD), and the Brunauer Emmett-Teller (BET). In addition, statistical analysis by the response surface methodology (RSM) was used to determine the optimum condition of the synthesis process in the range pH of 7.5 to 10.5 and the temperature range of 25°C to 65°C. The result suggests that the maximum catalyst mass was reached at precipitation pH 9 and 45°C. In this condition, the isomerization process obtained the highest yield and selectivity of 20.14% and 62.40%, respectively.

INTRODUCTION

The use of fructose in the food and beverage industry is currently widely practiced such as soft drinks [1], red wine of roselle [2], biscuits [3], kefir [4], and some of food and beverage products. The comparison degree of the sweetness (Dextrose Equivalent) from the glucose, sucrose and fructose are 74, 100, dan 180, respectively [5]. Based on the degree of sweetness, the use of fructose in small amounts will give a sweeter taste than glucose or sucrose. In general, fructose is obtained from the glucose isomerization process where the reaction is endothermic and reversible. This means that the maximum conversion rate of glucose into fructose can be achieved by adjusting the thermodynamic equilibrium between the two sugars

KEYWORDS

Temperature Humidity Screen house Internet of things Fan-pad evaporative cooling

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at a certain reaction temperature [6]. The proper isomerization process will produce fructose optimally.

Fructose production by enzymatic isomerization requires maintained conditions on the temperature and pH in order to the enzyme can work properly. This process required 36h to 43h [7,8] so it needed high energy consumption. For increasing the speed of the isomerization process and reducing the activation energy, several studies have been carried out using heterogeneous catalysts such as hydrotalcite [7,9]. The application of Mg/Al hydrotalcite catalyst is expected to provide a better performance in the isomerization process because it is easy to separate, can be used many times without losing the catalytic activity, and not contaminated easily [10].

The catalytic activity of Mg/Al hydrotalcite is determined from the surface area and its active site. The high active site of the catalyst will increase the interaction of substrate during the isomerization process and will increase the yield of fructose. Some parameters have to be considered during the synthesis of Mg/Al hydrotalcite as the catalyst such as pH and the temperature process. These two things will affect the surface area and crystallinity of the catalyst [11]. The common method of Mg/Al hydrotalcite synthesis is coprecipitation. This process is carried out under the saturated conditions by controlling the pH of the solution so that the deposition process of cation precursor can precipitate at the same time (in this case, Mg_2 + became M_2 + and Al_3 + became M_3 +). Inappropriate treatment of pH and temperature of precipitation will lead to the formation of poor crystals. High crystallinity can be achieved under supersaturated coprecipitation conditions in low concentrations [11]. Several studies have been conducted to determine the effect of operating conditions such as pH, temperature, and precipitation time during the synthesis process of Mg/Al hydrotalcite. Yu et al. [12] made Mg/Al hydrotalcite at room temperature in pH of 9.5 for 18 h and obtained the isomerization conversion, selectivity, and yield for the calcinated catalysts of 75%, 31%, and 23%, respectively. A higher result was obtained for the rehydrated Mg/Al hydrotalcite catalyst. Delidovich & Palkovits [10] carried out the synthesis of Mg/Al hydrotalcite for a longer precipitation time, 20 h, at the pH variation of 8.5 to 10.2 and a constant temperature of 30°C. It produced the highest isomerization yield of 30%. Yabushita et al [13] synthesized the Mg/Al hydrotalcite at the pH range of 7 to 8, and the precipitation temperature of 65°C. It was activated by the calcination and obtained 50% of isomerization yield. Yang et al [14] reported their study about the treatment of high precipitation temperatures during the catalyst synthesis obtained a good crystallinity but required high energy consumption. Mg/Al hydrotalcite catalyst made at the precipitation alkaline pH and temperature of 120°C for 1h produced a good crystallinity of the catalyst. The high crystallinity can also be achieved by adjusting the stirring speed on the optimal precipitation time [15]. The crystallinity of the catalyst will affect the catalytic activity [16,17]. Furthermore, The Mg/Al hydrotalcite catalyst should be checked for its catalytic activity by the isomerization process. In the previous study, Permanasari et al [7] reported their study about glucose isomerization of the onggok (solid waste of tapioca) with the comparison of Mg/Al hydrotalcite and glucose isomerase as the catalyst. The yield isomerization is 1.94% for the process with Mg/Al hydrotalcite in 7 h, and 6.18% in 43 h for the process with glucose isomerase. The Mg/Al hydrotalcite catalyst in this study was not activated by calcination and caused low yield isomerization. Based on these explanations, the effect of Mg/Al

hydrotalcite synthesis and their catalytic activity need to be studied further. The objectives of this study determine the effect of precipitation pH and temperature during the Mg/Al hydrotalcite synthesis on their catalytic activity on glucose isomerization.

METHODS

Chemicals and Reagents

The synthesis of Mg/Al hydrotalcite catalyst used magnesium nitrate hexahydrate (Mg(NO3)2.6H2O p.a.), aluminum nitrate nonahydrate (Al(NO3)3.9H2O p.a.), sodium hydroxide (NaOH p.a.), and Sodium carbonate (Na2CO3 p.a.) as a constituent chemical. We used pure D-glucose as the substrate and there are some reagents used for the analytical method. Resorcinol (p.a), thiourea (p.a.), and concentrated hydrochloric acid (p.a.) were used to analyze the fructose concentration while the sodium metabisulfite (p.a.), NaOH (p.a.), and 3,5-dinitro salicylic acid (p.a) was used for analytical glucose concentration. All the chemicals and reagents were provided by Merck, Himedia, and Sigma-Aldrich.

Determination of the Experimental Design and Statistical Analysis

The experimental design of the variable and response was determined by Response Surface Methodology (RSM) [18] to find out the effect of precipitation pH and temperature on the conversion, yield, and selectivity of the isomerization. The range of the pH used was 7.5 to 10.5, while the precipitation temperature was 25° C to 65° C. By the RSM, we got 13 experimental combinations of precipitation pH and temperature. The variable design of this study by RSM was shown in Table 1.

Run	pH	Temperature (°C)		
1	7.5	25		
2	10.5	25		
3	7.5	65		
4	10.5	65		
5	6.87	45		
6	11.12	45		
7	9	25		
8	9	73.3		
9	9	45		
10	9	45		
11	9	45		
12	9	45		
13	9	45		

Table 1. Varia	able design of the	precipitation	pH and tem	perature in Mg//	Al hydrotal	lcite synthesis
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Synthesis of Mg/Al Hydrotalcite Catalyst

Synthesis of Mg/Al hydrotalcite catalyst was made by coprecipitation method. The constant molar ratio of Mg/Al was set at 3:1 with a precipitation time of 18h. First, we made two

solutions in this process. The first solution contained Mg(NO3)2 and Al(NO3)3 according to the Mg/Al molar ratio set in the approximate volume of 200 ml. The second solution contained 0.1 N Na2CO3 and 1 N NaOH which served to control them according to the pH variation (see Table 1) to inserted CO32- as an interlayer on Mg/Al hydrotalcite with an approximate volume of 200 ml. These two solutions were placed in two different burettes and entered simultaneously into the flask drop by drop. We set the precipitation pH and temperature by the variation on the variable design (see Table 1). Mg/Al hydrotalcite was filtered by filter paper and washed to neutral condition (pH &) to remove the impurity of ions. We dried the Mg/Al hydrotalcite catalyst at 120°C to reach a constant weight and crushed it to a uniform size. The Mg/Al hydrotalcite catalyst was activated by the calcination process on the furnace at 500°C for 3h.

Characterization of Mg/Al Hydrotalcite Catalyst

We used Fourier Transform Infrared (FTIR), Ray Diffractometer (XRD), and Brunauer Emmett-Teller (BET) to characterize the Mg/Al hydrotalcite catalyst. FTIR, XRD, and BET are used to observe the functional group, the crystallinity of the catalyst form, and the specific surface area of the Mg/Al hydrotalcite catalyst, respectively.

Isomerization of Glucose to Fructose

Glucose isomerization into fructose was carried out to determine the catalytic activity of the Mg/Al hydrotalcite catalyst. This process was done in the 13 experiments according to the catalyst made. 10% (w/v) of D-Glucose solution became the substrate and was placed in a glass flask supported by an oil bath, a stirred apparatus, and a temperature sensor. We added 1% (w/v) of Mg/Al hydrotalcite catalyst for the 50ml substrate solution into the flask to carry out the process. Each process was set up at pH 7, 120 \Box C, for 1 h. The solution was cooled and analyzed the glucose and fructose concentration.

Analytical Method

The analytical method for determining the glucose and fructose concentration was DNS and seliwanoff's test, respectively. Seliwanoff's test is used to react the ketose group of the fructose with resorcinol and concentrated HCl. 2ml of the diluted samples were added by 7ml of HCl concentrate (5:1) and 1ml of resorcinol. The samples were heated the 80°C for 10 minutes. Then, they were cooled on the ice water to decrease the temperature suddenly. For analyzing the glucose concentration, DNS is used to react the aldose group in the glucose with the 3,5-dinitro salicylic acid reagent. 2ml of the samples were added by 3ml of DNS reagent. They were heated in the boiling water for 15 minutes and cooled in the ice water too. The spectrophotometry method was used to measure the absorbance of the samples. We used a visible spectrophotometer to detect the absorbance of ketose in the wavelength 520 nm, and aldose groups at 540 nm. Ketose groups represent the fructose concentrations were determined by comparing the sample's absorbance with the standard glucose and fructose

curves. Glucose and fructose concentration was used to calculate the yield, selectivity, and conversion. Therefore, the mass of catalyst product, conversion, yield, and selectivity were analyzed by RSM to see the effect of the precipitation pH and temperature on the isomerization process.

RESULTS AND DISCUSSION

The Mass of Catalyst Products

The Mg/Al hydrotalcite catalyst formed is a white crystal powder. It is the deposition of Mg2+ and Al3+. The molecule formula of hydrotalcite Mg/Al with the Mg/Al molar ratio of 3:1 is [Mg0.75Al0.25(OH)2] (CO3)0.125.0.62H2O(s) [19]. In this study, the precipitation pH and temperature did not affect the molecular formulation but affected the mass of the catalyst product.

The mass catalyst product of Mg/Al hydrotalcite in 13 experiments followed the variable design of the RSM was shown in Table 2. The smallest catalyst product was achieved in run 2 while the largest was in run 9, 1.97 g, and 4.52 g, respectively.

In the process of Mg/Al hydrotalcite synthesis, the too alkaline environment will cause Al₃+ ions in the Al(NO₃)_{3.9}H₂O to dissolve and will not depos with Mg₂+. An environment was potentially caused the Mg/Al hydrotalcite contain quite the impurities AlOOH or AlHO₂. Al₃+ ions which did not react with Mg₂+ caused decreasing of the Mg/Al molar ratio so which affected the mass catalyst product of Mg/Al hydrotalcite. The alkaline environment during the precipitation of Mg/Al hydrotalcite synthesis was should be maintained under at the neutral pH to pH 10 [17]. In this study, some experiments in run 2, run 4, and run 6 with pH over 10, was potentially contained many impurities on the Mg/Al catalyst formed.

Run	pH	Temp (°C)	Mass (g)	Yield (%)	Selectivity (%)
	X1	X2	Yı	Y2	Y ₃
1	7.5	25	2.81	3.97	8.90
2	10.5	25	1.97	4.85	13.39
3	7.5	65	2.39	4.11	16.45
4	10.5	65	2.63	6.91	26.89
5	6.87	45	2.28	3.77	16.21
6	11.12	45	3.13	3.80	7.10
7	9	25	2.55	11.62	38.23
8	9	73	2.47	13.52	41.53
9	9	45	4.52	20.15	62.40
10	9	45	3.01	13.49	34.25
11	9	45	4.16	15.02	44.18
12	9	45	3.05	15.16	40.96
13	9	45	3.84	16.31	43.37

Table 2. The central composite design of the experimental and the response

The precipitation temperature on the Mg/Al hydrotalcite synthesis played an important role to accelerate the motion of particles so that increased the rate of reaction. The heating process during precipitation increased the crystallinity of the Mg/Al hydrotalcite catalyst formed [11]. In this study, the optimal operating conditions of the Mg/Al hydrotalcite synthesis process obtained the largest of mass catalyst product was at 45°C and pH 9. Besides, the precipitation time had to be considered because it affected the Mg/Al hydrotalcite catalyst formation.

The Mg/Al Hydrotalcite formed is the mixing result of main and supporting ingredients. The crystal formed is a lyotropic crystal phase because it was formed by mixing two or more chemical compounds. The lyotropic phase is stable thermodynamically. It is stable when reacted with liquids and has very low toxicity. The alkaline conditions and low precipitation temperature during the synthesis of Mg/Al hydrotalcite cause the rhombohedral crystals form [11].

Catalytic Activity on the Isomerization Process

The hydrotalcite Mg/Al catalyst has been tested for its catalytic activity through the isomerization process. Table 2 shows the results of the catalytic activity test of Mg/Al hydrotalcite through conversion, yield, and isomerization selectivity.

The Mg/Al Hydrotalcite catalysts in run 9 and run 13 obtained a good catalytic activity based on the yield of fructose production, 20.15% and 16.31%, respectively. Yu et al [12] reported on their study about the isomerization with catalyst synthesized in pH 9.5 at room temperature obtained a slightly higher yield of 26%, while Steinbach et al [20] got a yield of 25% for 1h isomerization process. Even though the isomerization yield of this study was lower than Yu et al and Steinbach et al [12,20], it was better than the previous study about isomerization of onggok's hydrolysate into fructose, without calcination of Mg/Al hydrotalcite catalyst. It reached a yield of 1.94% [7]. In the previous study, Permanasari et al [7] used a hydrolysate sugar of Onggok for the substrate, while Steinbach et al [20] used a hydrolysate sugar of lignocellulose. The hydrolysate sugar as the isomerization substrate has more impurities than the pure sugar (glucose) so it needed a higher concentration to reach out higher yield. Besides, the selectivity of fructose became an important thing to be considered. Furthermore, the quality of the catalyst used is affected by the isomerization catalyst. The value of selectivity indicates isomerization quality. In this study, the highest selectivity was achieved at run 9 of 64.40%, followed by run 13 of 43.37%. The active site of the catalyst base will determine the direction of the isomerization reaction of glucose to fructose and may affect selectivity. Yu et al [12] described the glucose reaction pathways (isomerization and degradation) over a base catalyst (see Figure 1). Isomerization of glucose into fructose is favorable in the presence of a weak base site of the catalyst and at the low reaction temperature. On the other hand, the strong base site catalyst at high temperature will cause the glucose degraded into lactic acid, glyceric acid, glycolic acid, and formic acid. It was also reported that at specific pH and high glucose concentration, glucose can be converted to oligomeric acids in the presence of a strong base catalyst.



Figure 1. Glucose reaction pathway [12]



Figure 2. Surface plot of the precipitation pH and temperature effect to the catalysis performance (Yield and selectivity)

The catalyst quality affected to the isomerization of glucose into fructose. Table 2 showed that the precipitation of catalyst synthesis in pH 9 obtained the Mg/Al hydrotalcite with the best catalytic activity based on the isomerization yield and selectivity. Furthermore, the isomerization yield and selectivity were assessed as the response of the precipitation pH and temperature during the catalyst synthesis. It was illustrated by the surface plots on the RSM (see Figure 2). The isomerization yield and selectivity tend to increase from pH 6.8 to 9 along with increasing the isomerization temperature and decrease at the pH over 9 and 45°C. This study also reported that the catalyst made from the precipitation process at 90°C for 1 hour was considered effective to prevent degradation reactions and caramelization.

Statistical analysis by Response Surface Methodology

The correlation of precipitation pH and temperature to the mass of catalyst product, isomerization yield, and selectivity was analyzed by RSM. Besides the surface plot, analysis of variance was used to examine the significant effect of the independent variables (precipitation pH and temperature) on the dependent variables (mass catalyst product, yield, and selectivity).

The full quadratic approach was used to see the interaction of each and the combined variables. The P-value determined the significance of the independent variables to the dependent variables. It was compared with the significant value (α) which is generally used is 5% or 0.05. P-Value under or equal to α showed that the independent variable did not affect significantly the dependent variables and vice versa. Table 3 shows the P-value of each and combined variables interaction.

Table 3. The P-value of the dependent variable			
Independent Variables P-value of dependent variables			
	Y1	Y ₂	Y ₃
X1	0.719	0.605	0.941
X2	0.527	0.219	0.208
X1.X2	0.372	0.704	0.764

Based on the P-value, we can explain that in this study precipitation temperature and pH did not significantly affect the mass catalyst product, yield, conversion, and selectivity of the isomerization.

Characterization of Mg/Al Hydrotalcite Catalyst

The catalyst product of the lowest pH (run 5) and the highest pH (run 6) was selected to be characterized by FTIR. This selection based on the consideration of Mg/Al hydrotalcite precipitation at too alkaline environment will be dominated by the impurity oxides formation such as AlOOH. The characterization of catalyst using BET and XRD was carried out on two isomerization experiments that produced the largest yield of fructose (run 9 and run 13). The best catalytic activity of Hydrotalcite Mg/Al was achieved at these operating conditions.

FTIR spectra of run 5 and run 6 are shown with the wavenumber's references of each vibration in Table 4.

Table 4. The comparison of IR spectra on the Mg/Al hydrotalcite catalyst				
T TTL	Wavenumber (o	Wavenumber (cm ⁻¹)		
VIDPALION	Run 5	Run 6	Kei.	
Mg-O-Al	450	450	[19]	
Mg-Al	685	670	[19]	
NO3 ⁻	840	855	[19]	
C-C	1234.48	-	[21]	
CO32-	1383.01	1383.01	[21]	
C=O	1643.41 and 1705.13	1637.62	[21]	
C-H	2085.12 and 2424.60	-	[21]	
OH	3452.70	3468.13	[21]	

The bands at wavenumbers 685 cm-1 and 670 cm-1 indicate the presence of the Mg-Al vibration in both spectra [19]. The interlayer anion CO32- as a stabilizer of hydrotalcite Mg/Al is identified at a wavelength of 1383.01 cm-1 [20]. In addition, NO3- anions were identified at the wavenumber of 840 cm-1 and 855 cm-1. The presence of NO3- during the Mg/Al hydrotalcite synthesis indicates that CO32- have not completely displaced the presence of NO3-. Calcination

causes the formation of Mg and Al oxides. This is evidenced by the presence of Mg-O-Al vibrations in the FTIR spectra at a range of 480–440 cm-1 [19], [22]. The presence of these Mg and Al oxides will not affect the catalytic activity of Mg/Al hydrotalcite [16,23]. According to the similarity of these wavenumber bands and the references, we can claim that the catalyst made already has characteristics of Mg/Al hydrotalcite.



Figure 3. IR Spectra of calcined Mg /Al Hydrotalcite catalyst

The specific surface area becomes one of the important things that affected the catalytic activity of Mg/Al hydrotalcite since the active site of the catalyst is on it. We analyzed the specific surface area and the particle size by BET on the 2 samples, run 9 and 13 which generate the highest yield of isomerization. The result was shown in Table 5 with the comparative result from the other study.

Table 5. Comparison of specific surface area and the particle size				
	The Mg/Al hydrotalcite catalyst			
Mangurad payameter	Run 9	Run 13	Ref.	
measureu parameter			(Error! Reference source	
			not found. et al., 2016)	
Specific surface area (m ² /g)	106.48	112.15	154	
Particle size (nm)	56.35	53.50	61.1	

The Mg/Al hydrotalcite catalyst of this study obtained a better result from Moriyama's [24] because it had a small particle size with a large surface area. It was expected to be more effective as the catalyst to convert glucose into fructose. Several studies reported that synthesis of Mg/Al hydrotalcite at a higher pH, 9.5, gave a larger surface area [7,12,23], while the process was in neutral condition (pH 6-8) gave a smaller one [13]. The study about the pH effect to the specific surface area of Mg/Al hydrotalcite catalyst is necessary to develop for a narrower range. In addition, the specific surface area of the Mg/Al hydrotalcite catalyst was also influenced by the stirring process during the precipitation [15] and the activation process by calcination [25].

The activation process of Mg/Al hydrotalcite by calcination not only increases the surface area but also the crystallinity of the catalyst. Crystallinity or Degree of Crystallinity (DoC) refers to the degree of structural order of a solid. In a crystal, the arrangement of atoms or molecules is consistent and repetitive. Increasing the crystallinity will increase the catalyst stability and reduce the activation energy of the reaction. A crystalline catalyst provided a better reuse cycle than the amorphous catalysts [26]. We used XRD to analyze the crystallinity of this catalyst. Sharp peaks of the diffraction pattern indicate the catalyst has the crystalline form while the weak peaks indicate the amorphous form. The sample of runs 9 and 13 obtained the two highest isomerization yields from the 13 experiments. The diffraction pattern of the two catalyst samples can be seen in Figure 2.

By the diffraction pattern of XRD, we determined the crystal size of the catalyst by the Scherer equation in equation 1 and analyzed the Degree of Crystallinity (DoC) by Match 3 software.

$$d = \frac{k \lambda}{\beta Cos\theta} \tag{1}$$

d = crystal size

k = dimensionless shape factor, with a value close to unity (typically value of about 0.9)

- λ = wavelength of the X-ray
- β = line broadening at half the maximum intensity (FWHM), after subtracting the instrumental line broadening (in radians)

 θ = Bragg angle



Figure 4 Diffraction pattern of Mg/Al hydrotalcite in run 9 and run 13

This diffraction pattern also gives a prediction of the Mg/Al hydrotalcite relative content by identifying the peaks according to The Joint Committee on Powder Diffraction Standards (JCPDS) card 00-014-0191, ascribed to carbonated Mg-Al hydrotalcite. The percentage of the compound in the sample is obtained by comparing the relative intensity of the diffraction peaks because they are proportional to the fraction of the compound in the sample [27]. It is calculated by equation 2.

relative content of Mg/Al hydrotalcite(%) =
$$\frac{[I/I1]s}{[I/I1]t} \times 100\%$$
 (2)

[I/I1]s = The sum of the relative intensities of the compound peaks in the sample which compare with the JCPDS

[I/I1]t = The sum of relative intensity of the sample

The degree of crystallinity, crystal size, and percentage of relative content of Mg/Al hydrotalcite were shown in Table 6.

Table 6. Characteristics of the crystal form of Mg/Al hydrotalcite				
Samples	DoC (%)	Crystal Size (Å)	Relative content (%)	
Run 9	26.49	5.57	81.03	
Run 13	30.64	1.6	79.80	

Both of the samples have a slight difference in relative content since they have the same operating condition during the synthesis process. The catalytic activity of the catalyst might be influenced by the relative content of Mg/Al hydrotalcite and the crystallinity. Yang et al [14] made Mg/Al hydrotalcite with the Mg/Al molar ratio of 3:1 and obtained a semi-crystalline structure with a crystal size 3.048 Å, lower than the crystal size of catalyst in run 9. The higher relative content of Mg/Al hydrotalcite causes higher catalytic activity even though with a slightly lower DoC. The DoC showed that this catalyst is in semi-crystalline form. The low crystallinity of the crystal form is caused by the duration and speed of stirring during the precipitation and the temperature [15]. The amorphous structure of this catalyst causes the brittleness of the catalyst when it is used in the isomerization process.

CONCLUSION

The precipitation process in pH 9 with temperature 45°C obtained the highest mass catalyst product of 4.52 g and catalytic activity proofed by the isomerization yield and selectivity of 20.15% and 62.40%, respectively. From the statistical analysis using RSM, it can be concluded that the precipitation pH and temperature did not have a significant effect on the mass catalyst product of the Mg/Al hydrotalcite and its catalytic activity. The precipitation pH and temperature may affect the crystal form of Mg/Al hydrotalcite. For its function as a catalyst, Mg/Al should have a high crystallinity so that it is not brittle during the isomerization process.

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