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## Rietveld Refinement Analysis of Lampung Natural Zeolite Catalyst Impregnated *Fe* with Diffraction Method Using MAUD Software

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**Abstract**— This research characterizes the space group and R-phase crystal structure using the XRD method and Rietveld refinement. Rietveld improvements using the Material Analysis Using Diffraction (MAUD) software are acceptable if the  $R_{wp} < 15\%$  and  $Sig < 2\%$ . MAUD is a diffraction/reflectivity analysis program based primarily on the Rietveld method and here uses X-ray reflectivity data on materials. The intensity of the diffraction data of the powder sample is the appropriate reflection intensity so that the atomic structure of the crystalline material can be determined based on the Le bail characterization technique. The reflectance intensity of this Fe/Zeolite is  $R_w = 9.51\%$  and  $Sig = 1.76\%$ . XRD analysis on Lampung activated natural Zeolite before impregnation showed a monoclinic clinoptilolite crystal phase, which gives the lattice parameters on  $a \neq b \neq c$  axis, axis angle  $\alpha = \gamma = 90^\circ \neq \beta$ . The Fe/zeolite in this study is a natural zeolite from Lampung, which was activated and then impregnated with Fe. The Fe/Zeolite catalyst Rietveld Refinement results showed Nepheline's closest crystal phase, which gave the lattice parameters  $a = b \neq c$ , axis, angle  $\alpha = \beta = 90^\circ$ ;  $\gamma = 120^\circ$ , the shape is similar to the hexagonal crystal structure (HCP). These results can be used as an initial reference for the study of the crystallization diffraction pattern in the further development of this catalyst.

**Keywords**— Material; crystal; structure; clinoptilolite; nepheline.

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### I. INTRODUCTION

Indonesia has many volcanic areas and is a rocky area that has the potential to produce zeolites. The use of Zeolite to be utilized in various fields is currently growing rapidly. This makes zeolite modification research very attractive to researchers, especially for Indonesian zeolites [1]. Indonesian natural zeolites are generally clinoptilolites, especially the Lampung natural zeolites [2]. Generally, zeolites are written with the chemical formula of oxides or based on crystal cell units, as follows;  $M_2/nO \text{ Al}_2O_3 \cdot a.SiO_2 \cdot b.H_2O$  or  $M_c/n \{ (AlO_2)_c (SiO_2)_d \} \cdot b.H_2O$ . The letter n; is the metal valence, a silicate molecule, and b; water, being c, d is the amount of tetrahedral alumina and silica. The d/c is  $SiO_2/Al_2O$  ratio varies from 1- to 51. At this time there are about 40 types of

natural zeolites and computer calculations have predicted that millions of hypothetical zeolite structures are possible. So far, only 232 synthetic zeolites whose structures are known, so this has become a question for zeolite scientists; why only a small fraction of the possibilities are observed, even though it has great potential. This is called the "bottleneck problem," which keeps research into the structure of zeolites like a puzzle [3].

Schaller discovered the mineral in Hoodoo Mountain Wyoming, United States in 1923 and named it "clinoptilolite". Clinoptilolite has a 4-4-1 ( $T_{10}O_{20}$ ) complex structure, with two-channel sizes of  $0.35 \times 0.79 \text{ nm}$  and  $0.44 \times 0.30 \text{ nm}$ . It also has a Si/Al ratio of 4.0-5.1 with a  $K > Na > Ca > Mg$  ion content [4]. The stereotypical structure of the clinoptilolite phase is shown in Figure 1.

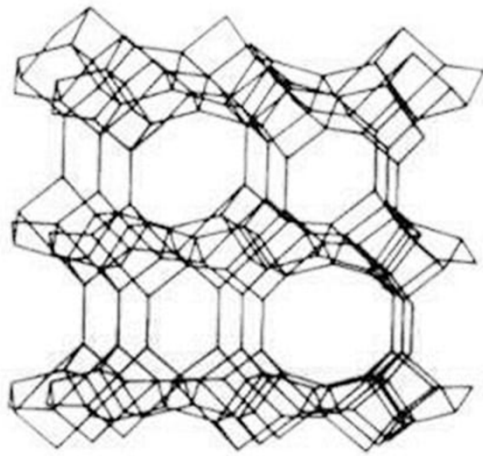


Fig. 1 The stereotypical structure of the clinoptilolite phase [5]

Zeolite identification is not only a chemical analysis but also an analysis of its structure is needed because Zeolite is used because of its unique structure. The uniqueness of the Zeolite structure lies in the structure-forming network that has a regular pattern with a certain cavity size. This results in the Zeolite being able to filter out certain reacting molecules. This capability is enhanced by making active site modifications, promoters, or adding dopants. Unique hollow structure and acidic properties make Zeolites widely used as catalysts in adsorption, alkylation, isomerization, and cracking [6], [7]. So it is necessary to analyze the crystal structure to determine the shape and properties so that it is easy to make and provides many benefits [8], [9].

Crystal structure analysis generally uses the XRD (X-Ray Diffraction) method. XRD is an analytical instrument commonly used to determine the characteristics of the crystal structure of a material by using the X-ray diffraction principle, known as Bragg's law, although X-ray diffraction was first discovered by Max von Laue in 1913<sup>th</sup>. Bragg formulated the diffraction process in equation mathematics as a result of the interaction between X-rays reflected by a material. This reflection does not cause energy loss but is in the form of elastic scattering [10]. The events described by Bragg show that the plane containing the atoms in the crystal when exposed to X-rays will reflect the same radiation as the light reflected in the mirror plane. The result of this reflection is a distinctive diffraction pattern. When viewed from the mathematical formula Bragg  $n\lambda = 2.dhkl . \sin\theta$ , where  $n$ ; is an integer,  $\lambda$ ; X-ray wavelength,  $dhkl$ ; distance between fields and  $\theta$ ; The angle of incidence of the X rays shows that two variables can be varied to produce a diffraction pattern, namely the wavelength and the diffraction angle. While the value of  $d$  is the edge that connects the crystal planes and has a fixed value for each particular crystal system [11].

However, if the crystal structure changes (for example, there is an interstitial process or an infiltration of the composite material) the value of  $d$  changes. The diffraction pattern generated from the XRD instrument can determine a crystal structure model [12]. The XRD test result is a curve depicting peaks in the diffraction pattern. Where the height of the diffraction peaks is a representation of the abundance of a crystal phase. The difference in the position of the atom/ion in the unit cell is indicated by the difference in height between

one peak and another. This difference in peak height is also the orientation of the crystal growth [13].

Rietveld was developed by Hugo Rietveld, who performs curve matching of calculated diffraction patterns (models) obtained from material crystallographic information related to the measured diffraction pattern [14]. In 1969, Rietveld demonstrated the possibility of replicating the measurement of diffraction patterns with pattern calculations. Pattern calculations provide an advantage in case of errors caused by intensity deviations in the imperfect model structure; they will tend to leave a lingering negative and positive intensity. The principle of Rietveld analysis is to match the calculated peak profile with the observed peak profile. This installation is done by applying a non-linear least squares calculation procedure [15]. Johnson et al. [16] performed Rietveld refinement of XRD data for several types of clinoptilolite zeolite and obtained a framework structure form; the simulation results were entered into the Crystallography Open Database known as CIF 1532909. However, there was no explanation of the simulation program used; it only is stated using the canonical Monte Carlo program [16]. Likewise, Tait et al. [17] with crystallographic information file 9004731, a nepheline phase for Canadian mineralogist crystals type known Fe-Clino.

Several authors who have carried out a structural analysis using MAUD software, such as Anand et al. [18], have conducted Structural and Vibration Research Improvements. The study confirmed that the Type M BaFe12O19 nanoparticles, XRD diffraction, that the ferrite samples were in a hexagonal structure with the P63/mmc space group. A phase characterization has been studied in the form of memory alloy Fe-Mn-Si-Cr-Ni which was processed by a thermomechanical method whose different phases were found to be isostructural with the  $\pi$  Cr3Ni5Si2 phase with the P213 space group. Vershinina Boev, and Ivanov [19], New Mo10Ni3C3B phase crystal structure; refinement shows that the new phase has a space group P63/mmc (Pearson symbol hP34) and lattice parameters  $a=7.8089$ ,  $c=7.8712$ . Naji, Khalil-Allafi, and Khalili [20] revealed that microstructural characterization and quantitative phase analysis of Ni-rich NiTi after stress-assisted aging for a long time using the Rietveld method [20]. Hadeef and Ans [21] performed a Rietveld refinement of the diffraction data on ball-milled Fe50Al35Ni15 powder, which stated that the ball-milled had a BCC phase. Saville et al [22], Rietveld refinement software MAUD for evaluating the crystallographic texture of single- and dual-phase materials, as applied to High-Pressure-Preferred-Orientation (HIPPO) neutron diffraction data obtained at Los Alamos National Laboratory (LANL) and electron backscatter diffraction (EBSD) pole figures on Ti-6Al-4V produced by additive manufacturing [22].

This process is based on the least-squares method which is then used as the basis for the algorithm. Some of the software commonly used for material analysis are Rietica [23], MAUD [24], and FullProf [25]. The Rietveld analysis used MAUD (Material Analysis Using Diffraction) software in this study. MAUD software is considered the most precise in analyzing material behavior [22].

MAUD is a software that uses JAVA language, which is very special in analyzing crystal structures based on diffraction data. Another feature that can be extended from

MAUD is the diffraction data file format. So that Maud recognized and loaded the new data format it was possible to compile the simulation results and add them to the library's crystallography information file [22]. Peak function computing, a special Java native vector library (in C/C++ language) including Pseudo-Voigt which has been developed for Altivec looks difficult to facilitate by MAUD software, so it can be said to be user friendly for reading diffraction patterns on crystallography [22], [26].

The author has synthesized several catalysts for the cracking process [27], and we had found a catalyst that is simple and has high selectivity in the formation of biogasoline, but the yield produced relatively small, that catalyst is Fe/Zeolite natural of Lampung [28]. So that we are interested in studying the structure of the Fe/Zeolite catalyst by improving the catalyst by analysis of the Rietveld refinement of XRD data.

The purpose of this study was to obtain an overview of the structure of the Fe/Zeolite catalyst as an initial reference for the further development of this catalyst. The results of this refinement of Rietveld are expected to enrich the Crystallographic Information Framework (Cif) for various modified Zeolites.

## II. MATERIAL AND METHOD

### A. Materials

Natural Zeolite is the main ingredient in this research which was obtained from PT. Winatama of Lampung. This zeolite material is the main material used as a support which will be impregnated with Fe metal from  $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ . This experiment also uses additives such as  $\text{NH}_4\text{Cl}$  and  $\text{AgNO}_3$  where all these chemicals are pure analytical grade obtained from PT. Merck Indonesia.

### B. Instrumentation

This research uses equipment in the form of laboratory glassware and a thermometer. To achieve the analysis using an XRD instrument (X-Ray Diffractometer Shimadzu), whose test results data will be analyzed with MAUD software (Material Analysis Using Diffraction New Version 2.99) to obtain the objectives of this study. Then the TGA instrument (Thermogravimetry Analysis Linseys) strengthens the results of the analysis.

### C. Procedure

1) *Natural Zeolite Activation:* The natural Zeolite of Lampung was sifted into 100 mesh then soaked in aqua distillate while stirring, let stand and continue the next day for 3 days (every day for 8 hours) at room temperature. Then filtered and the clean precipitate was dried in an oven at  $120^\circ\text{C}$  for 2 hours. Then the Zeolite was immersed in 1 M  $\text{NH}_4\text{Cl}$  solution for 24 hours at room temperature. After completion, the Zeolite was filtered and washed with aqua distillate and tested for chloride ion ( $\text{Cl}^-$ ) content using silver nitrate ( $\text{AgNO}_3$ ) solution until it doesn't contain  $\text{Cl}^-$  anymore. Furthermore, the Zeolite was dried in an oven at a temperature of  $120^\circ\text{C}$ . After cooling, the Zeolite was placed in a porcelain dish and calcined overnight, at  $450^\circ\text{C}$  in a furnace. Then cooled at room temperature, and obtained active Zeolite (H-zeolite) which is ready to be impregnated with  $\text{Fe}^{3+}$  [2].

2) *Impregnation of the catalyst with Fe:* The H-zeolite was impregnated with a solution containing the active metal, namely  $\text{Fe}(\text{NO}_3)_3$ , by slowly adding the metal solution to the H-zeolite at room temperature. The solid was then dried at  $110^\circ\text{C}$  for 14 hours, then calcined at  $500^\circ\text{C}$  for 4 hours [28].

3) *Analyzed using XRD and Rietveld Refinement XRD Data with Material Analysis Using Diffraction (MAUD):* Smoothed sample and inserted into the sample holder appropriately, the surface homogenized, or flattened, then placed in the measurement position. Measurements were carried out by the operating system of the XRD Shimadzu Type 7000, Cu  $\text{K}\alpha$  radiation. The diffraction pattern was scanned from  $10$ - $90^\circ$ ,  $2\theta$ . The speed used is  $2^\circ/\text{min}$  in continuous and absolute methods. The tube voltages and tube currents of the X-ray generator are 40kV and 30mA [29].

This XRD instrument has software that records the results of the XRD spectrum into a data and diffraction pattern curve using the standard from the International Center for Diffraction Data (ICDD 53-1178), with a measurement time of 45 minutes for each sample [2]. The resulting diffraction patterns were analyzed in Rietveld using MAUD software. By using Cif (Crystallographic Information File) from COD (Crystallography Open Database). Selection of Cif based on references by the results of sample analysis with XRD. A simulation is carried out to find the closest to refinement [30], [31].

Quantitative estimation of crystallographic defects such as stacking faults, twinning, and dislocation density is very likely to be analyzed by MAUD software. MAUD has the main features Written in Java can run on Windows, MacOSX, Linux, and Unix (need Java VM 1.6 or later). MAUD is easy to use, every action is controlled by a GUI. Works with X-ray data, synchrotron, Neutron, TOF, electrons developed for Rietveld analysis. Supported Ab-initio structure solution integration, from peak finding, and indexing to solving Different optimization algorithms available (LS, Evolutionary, Simulated Annealing, Meta dynamics). Analysis used Le bail fitting. Several data files input formats Works and input images from 2D detectors (image plates, CCD). CIF trials compliance for input/output; import structures from databases (Crystallography Open Database / COD) [24].

4) *Analyzed thermal test using TGA:* The thermal test on the Fe/Zeolite catalyst used Thermo Gravimetric Analysis (TGA). In TGA, changes in sample weight as a function of temperature and time are recorded automatically. Nitrogen gas with a flow rate of 40 mL/minute flows into the TGA furnace and gas with a flow rate of 60 mL/minute. Then the sample weighing 30-40 mg is put into a platinum container in the furnace. The temperature is increased at a rate of  $10^\circ\text{C}/\text{minute}$ . minutes and for 100 minutes to  $1000^\circ\text{C}$ . Measurements of the mass fraction of the sample during the experiment were recorded [27].

In this thermal analysis, the mass of the sample that changes due to the decomposition, adsorption, or reaction processes is monitored as a function of temperature.

The scheme of this research is shown in Fig. 2.

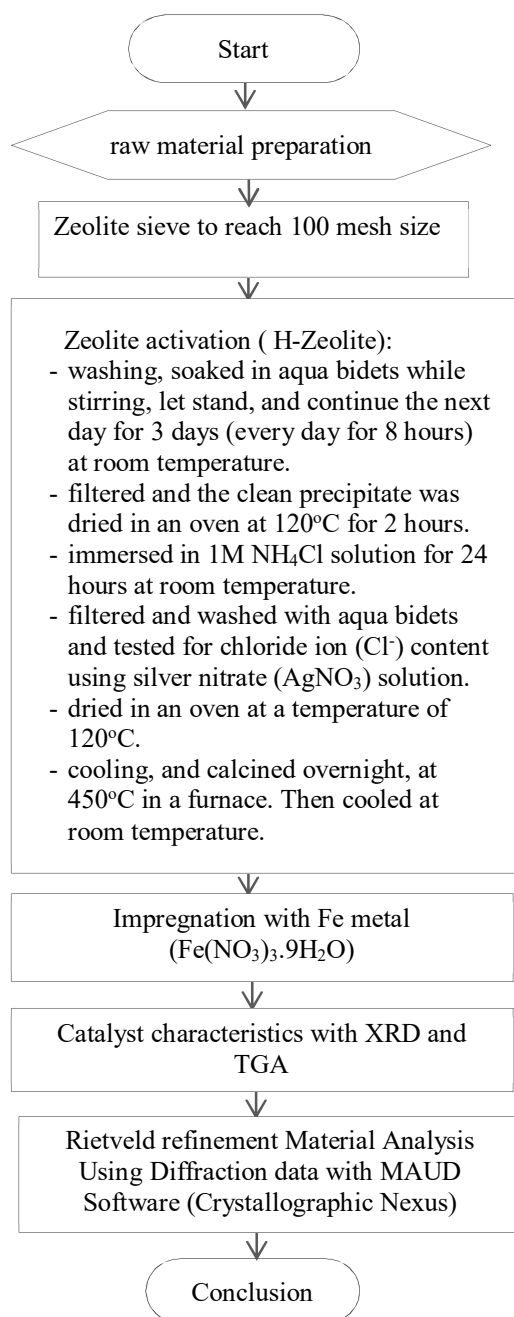


Fig. 2 The scheme of research

### III. RESULTS AND DISCUSSION

The analysis carried out is a quantitative phase analysis of the XRD Fe/zeolite data from Lampung. XRD test gives a diffraction pattern. On this diffraction pattern, a refinement was carried out using MAUD software. The diffraction pattern of Fe/zeolite from Lampung XRD test results is shown in Fig. 3. That are 3 (three) highest peaks from the sample (Fe/Zeolite) at an angle of  $2\theta$ , as shown in Table 1, that is  $28.0113^\circ$ ,  $22.3735^\circ$ , and  $9.8590^\circ$ . Natural Zeolite of Lampung is known to have a type of clinoptilolite which generally has a peak of 22, 28, 30, [2], so it can be seen that there are differences in peaks after impregnation Fe metal. this is explained in fig. 4

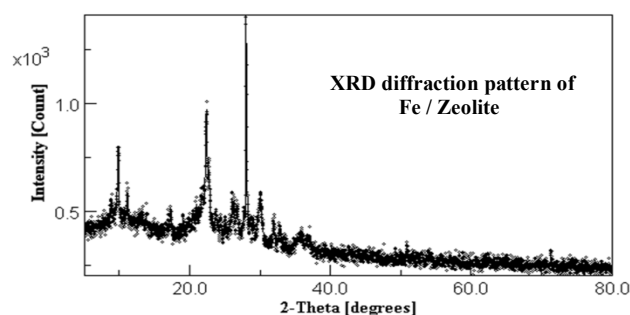


Fig.3 XRD diffraction pattern of Fe / Zeolite sample

TABLE I  
THE HIGHEST PEAK OF THE FE / ZEOLITE DIFFRACTION PATTERN

# Strongest 3 peaks

No	Peak no	2Theta (deg)	d(A)	I/I1	FWHM (deg)	Intensity (Counts)	Integrated Int (Counts)
1	31	28.0113	3.18283	100	0.10870	805	5532
2	20	22.3735	3.97047	42	0.26160	342	4559
3	4	9.8590	8.96428	29	0.19190	231	2958

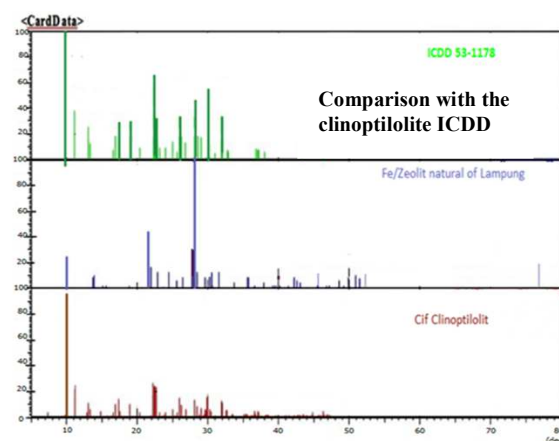


Fig. 4 Comparison with the clinoptilolite data card from the ICDD data card.

Fig. 4, is the results compared with the existing clinoptilolite data card and showed that the zeolite catalyst impregnated with Fe (Fe/Zeolite catalyst), has a significant similarity to the clinoptilolite type crystal;  $71-1425 \text{ Na}_4.12\text{Si}_36\text{O}_72 (\text{H}_2\text{O})_{23.12}$ . [2], [32]. but it can also be said to be similar to type zeolites in the area where Agoura, USA is included as clinoptilolite type zeolite, HEU type structure with chemical composition;  $(\text{K}1.76\text{Na}1.84\text{Ca}1.24\text{MgO}.20) (\text{Si}29.84\text{Al}6.16\text{O}72).21.36\text{H}_2\text{O}$  [2]. With space group symmetry;  $C12/m1$ , crystal system: Monoclinic, lattice parameters:  $a = 17.662$ ,  $b = 17.911$ ,  $c = 7.407 \text{ \AA}$ , angles between axes:  $\alpha = 90^\circ$ ,  $\beta = 116.4$ ,  $\gamma = 90.0^\circ$  [33].

Then the XRD test results data were subjected to a refinement of the clinoptilolite type coordinates CIF 1532909 from COD (Crystallography Open Database). The simulation is carried out by analyzing the crystal structure program in MAUD (Material Analysis Using Diffraction) software. The simulation process is run by entering intensity data from the XRD test on the sample (Fe/Zeolite catalyst), along with structural parameters (CIF). MAUD software extracts measured intensity data using nonlinear least-squares fitting by Le Bail. Le Bail extracts the intensity (I, hkl) from the

powder sample diffraction data to find the appropriate intensity so that the atomic structure of the crystalline material can be determined. This technique is also for repairing unit cells and has the added advantage of checking phase purity [23]. Le Bail on MAUD can also fix the intensity of the overlapping diffraction data with a similar distanced. This is because the intensity is allocated based on the number of intensities that contribute to a particular peak [22].

Refinement is performed using an algorithm in software that involves purification of unit cells, profile parameters, and peak intensity so that they can be adjusted to the measured powder diffraction pattern. Le Bail can also be used to find phase transitions in high pressure and temperature experiments. Le Bail analysis is believed to be a method that can provide an estimate for the intensity of the reflection allowed at various crystal symmetries. Thus, the Le Bail technique is especially relevant for diffraction studies that involve the use of a radiation source in the form of a neutron or a. Le Bail's work initially found the peak positions in the data, then indexed patterns, which were then used to define unit cells or lattice parameters [34].

The determination of the space group follows based on symmetry and the presence or absence of certain reflections. In modified or engineered crystals, usually at the beginning of the Rietveld provide a measured value which will result in a bias with the calculated value. Refinement by Le Bail by assigning the new calculated structure factor to the new structure factor value. At this point, refinement is performed on the unit cell, background, peak width, peak shape, resolution function, and increases the parameters. The structure factor is then reset to the new structure factor value and the software starts the process again, according to the iteration performed. The success of this refinement is of course in addition to the use of the right software as well as the ability to select the initial computed data structure (CIF). That pattern decomposition programs based on the Le Bail algorithm can exploit the prior information [34], [35].

The selected CIF based on reference analysis refers to the type of catalyst carrier, namely the clinoptilolite type of natural Zeolite Lampung. CIF shows the room group symmetry. Fig. 5 shows the symmetry of this space group with the Hermann maugin space group symmetry C 12/m 1 for clinoptilolite. It can be said that the crystal system is monoclinic, seen also from the lattice parameter (Bravais unit cell) axes  $a \neq b \neq c$  and axis angles  $\alpha = \gamma = 90^\circ \neq \beta$ .

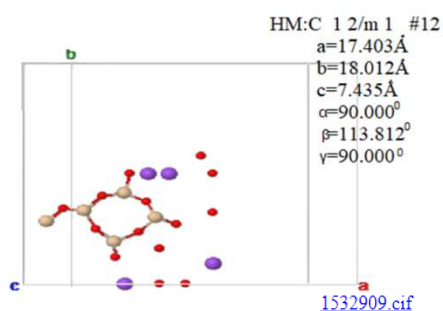


Fig. 5 The spatial symmetry of the CIF coordinate

Refinement of sample XRD intensity data with Cif 1532909 shows the intensity as shown in Figure 6.

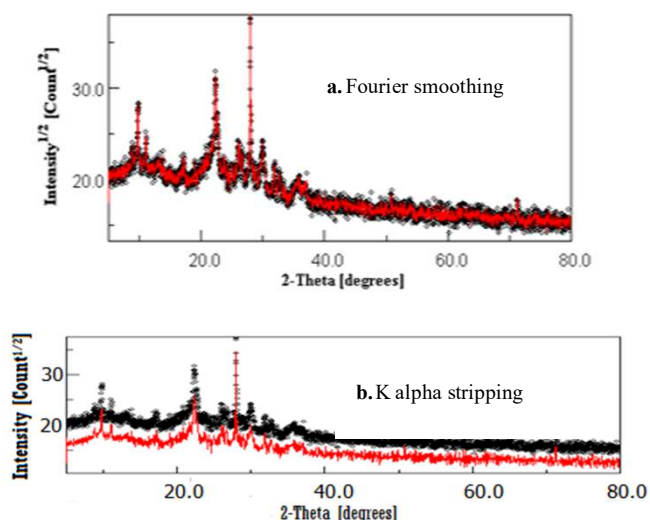


Fig. 6 Rietveld refinement of MAUD software. The measured diffraction pattern is depicted in black. The calculated diffraction pattern is depicted in red. a. result Rietveld refinement with Fourier smoothing. and b. result Rietveld refinement with K alpha stripping

The Refinement results CIF 1532909 can be seen in Fig. 6, which states that XRD data similar as clinoptilolite. Fig. 4a. Fourier smoothing, and 4b. K alpha stripping showed a small difference in peaks between XRD and CIF data. This K alpha stripping image illustrates that the peaks show a dual structure, caused by the K1 and K2 emission lines from the X-ray source, showing information about the difference wavelength, intensity, and shape of the K1 and K2 lines. This difference in wavelength there is different because the XRD data is not pure clinoptilolite but has been impregnated by Fe metal.

Figure 6 also shows that the results of refinement produce Sig (Sigma Values) 1.46% and Rwp (R-weighted profile) 7.86%, which can be said to be in the successful refinement category [36], [37]. Rietveld's results also show that clinoptilolite with CIF 1532909 has an isometric system known as cubic crystal, the uniqueness of the Lampung natural Zeolite has an isometric order structure with Al and Si atoms, so it can be said to have a cubic crystal structure of diamond, with cell volume  $2132.2A^3$ . This crystal also has optical isotropy, meaning that it has the same optical properties in all directions. The crystal system is described not only as geometry but more in terms of the arrangement of atoms and unit cells.

This refinement was carried out on CIF Fe, to see the intensity of the Fe metal that was carried. The results of the MAUD refinement are as follows:

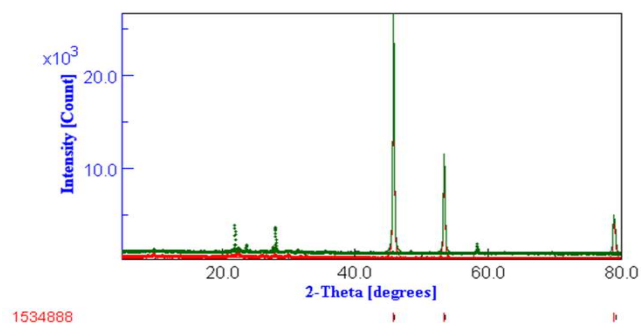


Fig. 7 Rietveld refinement sample of Fe / Zeolite with Cif Fe 1534888

Fig. 7. is a Rietveld of XRD test results for Fe/Zelite catalyst with Cif Fe 1534888 showing that the sample is estimated to contain impregnated iron in the natural Zeolite of Lampung support. shown with a peak at  $2\theta$ ; at 45.7817, 53.3786 and 78.8692.

Rietveld Refinement analysis of Fe/Zelite catalyst with CIF which is estimated to be more representative than the results of the previous analysis, using CIF clino-Fe. Where clinoptilolite containing Fe is considered to be almost the same as the Zeolite type by Canadian mineralogists[38]. Canadian mineralogist symmetry is seen from the data in Fig. 8 is P 63, with lattice parameters  $a = b \neq c$ , axis angle  $\alpha = \beta = 90^\circ$ ;  $\gamma = 120^\circ$ . It can be said to have a Hexagonal crystal system, with a cell volume of 725,875  $\text{Å}^3$ .

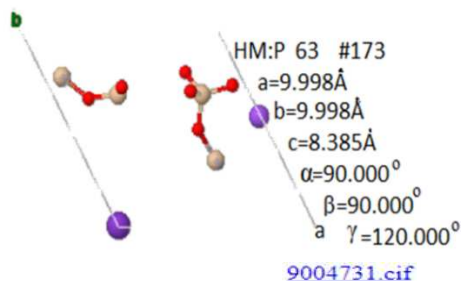


Fig. 8 Coordinates of cif

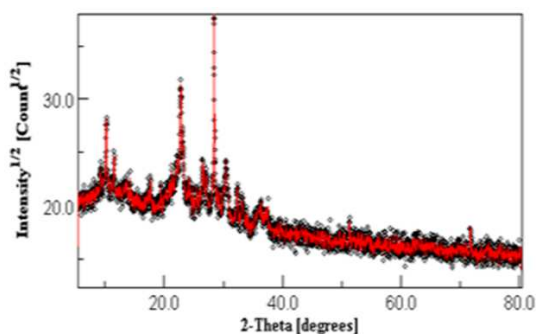


Fig. 9 Rietveld refinement, measured data (black color) with Cif (red color) using MAUD software.

In Fig. 9, we can see the results of the analysis of Rietveld refinement Lampung natural Zeolite catalysts with impregnated Fe metal using CIF Clino-Fe. Fig. 8 shows the suitability of the measured diffraction pattern from the XRD sample test results with those calculated from crystallography open database. Based on the analysis of the activated Lampung natural Zeolite that unimpregnated Fe has a Clinoptilolite phase with a Monoclinic crystal system, which provides lattice parameters for  $a \neq b \neq c$  axis, axis angle  $\alpha = \gamma = 90^\circ \neq \beta$  with a density of 1.7898. Isometric as well as isotropic properties make the crystal can be said to have the cubic structure of diamond. This is because it has the same 2 (two) lattice angles, and a crystal structure like this is usually referred to as an AX type mixed crystal structure, where A is the notation for cations while X is for anions. The cubic structure of diamond has 8 (eight) atoms occupying an angular position which is one-eighth of the size, so it is said that the coordination number of the cubic diamond structure is 4 [39].

Therefore, the zeolite catalyst that has been impregnated with Fe metal, based on the results of the Rietveld refinement analysis is similar to the type of Canadian mineralogist

crystals, called Nepheline [40]. Nepheline has a Hexagonal crystal system seen from the lattice parameters of the axis  $a = a \neq c$ , the axis angle  $\alpha = \beta = 90^\circ$ ;  $\gamma = 120^\circ$ , this change is due to the clinoptilolite being exposed to Fe metal.

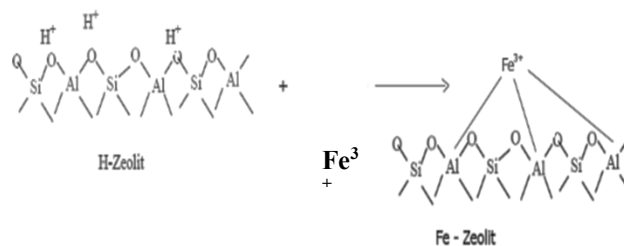


Fig. 10 Fe Impregnation reaction

Reaction in fig. 10 shows impregnation Fe in the Lampung Natural zeolite changes the position of the alumina silicate in the crystal core and makes Fe the active site. This process is similar to the solidification process of cast Fe, which causes an increase in density. From the results of Rietveld refinement, the density of the catalyst Fe/Zelite was 3.7174. Quantitative estimation of crystallographic defects such as stacking faults, twinning, and dislocation density is very likely to be analyzed by MAUD software.

This increased density results in the strength of the catalyst to heat, making it particularly suitable for the cracking process. It can be seen from the curve of the TGA (Thermo Gravimetry Analysis) test.

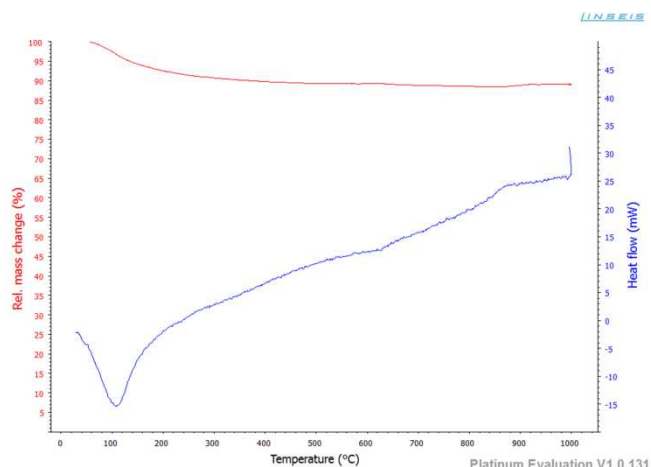


Fig. 11 Curve of the TGA test results for Fe/Zelite catalyst

Fig. 11 shows a heat-stable Fe/Zelite catalyst. The mass decomposition occurs at  $61^\circ\text{C}$ , most likely due to the loss of volatile elements. The blue line on the curve shows that the reaction takes place in atmospheric conditions with good stability to heat and the reaction is exothermic.

Lampung natural Zeolite catalyst impregnated with Fe from the results of Rietveld refinement shows that the modified catalyst in principle retains the properties of clinoptilolite, but the modification causes the crystal shape to lead to Nepheline. This may be the reason why Fe impregnated Zeolite catalyst has high selectivity for the hydrocarbon catalytic cracking process [28]. In addition, Fe has magnetic properties that help in the more selective termination of the hydrocarbon chain[36]. FoM (Figure of



Merit) Rietveld Refinement MAUD provides criteria such as Table 1.

TABLE II  
FIGURE OF MERIT (FOM) VALUES OF REFINEMENT USING MAUD SOFTWARE

Fe/Zeolit	FoM (%)				
	<i>Sig</i>	<i>R<sub>w</sub></i>	<i>R<sub>wp</sub></i>	<i>R<sub>b</sub></i>	<i>R<sub>exp</sub></i>
	1.76	10.16	9.51	6.99	5.39

The criteria for the re-level Refinement results in table 2 show the fulfillment of the Refinement requirements, are the value of  $R_{wp}$  9.51% <15%, where  $R_{wp}$  is the R index associated with the sum of the residual squared weights.

The best solution found for refinement in most programs is the value of  $R_{wp}$ .  $R_{wp}$  is Rietveld-like R factor or "intensity-weighted/weighted profile- which takes into account the normal Gaussian distribution standard deviation for each measured intensity[18]. It is a better indicator of the orientation distribution refinement reliability for comparing different samples.

The value of  $Sig$  (signal value) is the difference between the measured and calculated intensity and this value is often used to represent the value of  $Gof$  (Godnes of fit). The  $Sig$  value which indicates a good refinement quality is <2% [41].  $Gof$  is a value that shows how well the results of the fitting are against the observation, namely the XRD test results[35].  $Gof$  is a value commonly used in statistics to describe the success of a fitting or simulation. Rietveld with some software refinement  $Gof$  measure is often expressed as the parameter value  $\chi^2$  (chi-square goodness fit). The  $Sig$  value obtained from the results of Rietveld Refinement of natural Fe / Zeolite catalyst in Lampung with MAUD software is 1.76%, so it is stated that the refinement quality is acceptable.

This phase change may have caused the natural Zeolite of Lampung impregnated with Fe to have high selectivity as a catalyst in producing biofuel from the catalytic cracking process of used cooking oil [28]. However, the yield which is not as expected has become the main attraction for conducting further research based on the results of this study to produce better modifications [42].

#### IV. CONCLUSION

Data Fe/Zeolite with MAUD software has been used in this research work for detailed structural analysis. The main conclusions of this experiment can be stated that the impregnation of Fe into natural Zeolite of Lampung causes the formation of the Nepheline phase with Hexagonal crystal form, which gives the lattice parameters  $a=b \neq c$ , axis angle  $\alpha=\beta=90^\circ$ ;  $\gamma=120^\circ$  and Hexagonal Closed Packed (HCP) crystal structure. Fe as a cation in Lampung natural Zeolite catalyst forms ionic bonds.

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