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Effects of Composition and Particle Size of Crystallization on Physical Properties of Marble Composite

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Abstract

Composited tile was made using marble particles, phenol resin and hexamethylenetetramine (HEXA) as the catalyst. The matrix and hardener of these materials were mixed on volume variation from 62.50 ml to 125.00 ml, and on variation of 25, 40 and 60 mesh. Samples were dried in a room temperature for 3 hours. The compressive strength and crystal structure were analysed. The results showed that compressive strength values were in range of 6.15×10^7 N/m^2 – 9.61×10^7 N/m^2, and there were two crystal structures consisted of marble crystal and phenol crystal. The analysis was carried out by using the Rietveld semiquantitative analysis. The final crystal structure of marble is rhombohedral, where the lattice constants are $a = b = 4.969$ Å; $c = 17.026$ Å. This results indicate that the composites and marble can be used to make tile, because they were lighter, stronger and the amount of crystals increases compared with the pure marble.

***Keywords** : Particle Size; Composite Material; XRD; Rietveld Analysis.*

1. INTRODUCTION

The growth of construction industry in Indonesia and Italy has developed so rapidly that the researcher try to get a new alternative construction material having better quality. They should be lighter, stronger, long lasting, and relatively cheaper compared with conventional one. This composite material is a type of construction material that consists of mixture or combination of two or more substances so that it acquires better quality than the parts of its components [1]. Marble is one of the commodities having good marketing prospect domestically and abroad since it deals with people's primary needs for construction material such as tile, wall, bill board and for electric isolator; home utensils such as table, cup, and there are some another things [2]. Marble polishing is carried out in order to get smooth, white or even having a certain colour pattern of marble. Furthermore, it produces waste products as tiny particles or marble fraction, because of developing of marble industry makes marble waste products will increase [3]. The waste products can be use for making composite tile as an alternative construction material.

In this paper we report on the fabrication of marble composite by drying process in room temperature. The crystal structure and the physical properties of the marble composite related to the composition and particle size are described.

2. EXPERIMENTAL

The particles of marble were obtained as the waste products of marble tile making in Citatah West Java Indonesia and furthermore were cleaned from dirt and dust with a paint brush. They were ground by a mortar, and refined using Nurmit Schutzring Machine. They were sieved with a 25, 40 and 60 mesh to get finer particles of marble. The sample specification is shown in table 1. To fabricate the sample first marble particles were poured into phenol resin and it was stirred during the process, the catalyst hexamethylenetetramine (HEXA) was added. The mixture was then poured onto 5 cm × 5 cm and 5 cm × 25 cm matrix and they were dried in room temperature for 3 hours. To get clean sample the residual mixture was removed. The compressive strength of the sample was characterised by Universal testing machine [4,5].

The crystal structure was analysed by x-ray diffraction (XRD) and Rietveld refinement. The XRD spectra were recorded on a Shimadzu type 610 diffractometer using CuKα radiation at 30 kV and 30 mA (900 watt).

Table 1.. Composition of sample composite

Sample code	Sample size		Particle of marble (ml)	Phenol resin (ml)	Catalyst HEXA (ml)
	Mesh	µm			
M125	25	710	62.50	62.50	0.50
M225	25	710	93.75	62.50	0.50
M325	25	710	125.00	62.50	0.50
M140	40	425	62.50	62.50	0.50
M240	40	425	93.75	62.50	0.50
M340	40	425	125.00	62.50	0.50
M160	60	250	62.50	62.50	0.50
M260	60	250	93.75	62.50	0.50
M360	60	250	125.00	62.50	0.50

Note in the first column :
M125 means sample with composition 1, that is 50% of 25 mesh of marble particle
M225 means sample with composition 2, that is 60% of 25 mesh of marble particle
M325 means sample with composition 3, that is 67% of 25 mesh of marble particle and so on.

3. RESULTS AND DISCUSSION

A. Compressive Strength Measurement

The result of the compressive strength measurement can be seen in Figure 1 and 2. They show that in increasing the fraction of the marble particle volume will increase the compressive strength of the composite [1], and has also shown that the marble particle refining will increase the value its compressive strength [6]. The compressive strength value of M260 (composition 2, 60 mesh) is 9.61 x 10^7 N/m^2, this value is lighter than 8.00x10^7 N/m^2 as the compressive strength value of a tile [7]. This is probably caused by the fact that there is not yet any proper contact on the surface between marble particle and phenol, as indicated by the presence of the pores. Based on the measurement, the sample with composition 2 has higher value of compressive strength than those of composition 1 and 3. This is assumed that the composition 2 gets optimum proportion between marble particle and its resin, so that in this composition there is a strong unity between marble particle and its resin in another word there is a proper surface contact. When the sample given the burden, the burden will be transferred to the whole area. This is also seen from the fraction pattern for the composition 2 showing fraction pattern which is more crystals than another composition, it means the this material is stronger than another.

B. Crystal Structure Measurement

Figure 3 shows XRD spectra of marble composite for various composition. The presence of intense diffraction peak corresponding to (104) plane implied that the marble composite assessed a strong preferential orientation. Crystal structure analysis was carried out by using the Rietveld semiquantitative analysis.

The Rietveld refinement [8] was carried out using the program PCRTVD [9,10]. The structure of Java's marble is formed by calcium carbonate ($CaCO_3$) [5,11,12], space group $R\bar{3}C$, was used as the initial model of $CaCO_3$. A search in the ICDD-PDF database [13] using the software available with the diffractometer was identified : $CaCO_3$ (PDF N° 24-0027). The peak positions of each phase were extracted by means of single-peak-profile-fittings. The remaining 12 intense peaks corresponding to the phase of interest, $CaCO_3$, were readily indexed in a rhombohedral cell. Table 1 contain the observed and calculated X-ray powder diffraction data for $CaCO_3$

The final refinement involved 23 parameters and 20 iterations, including two theta zero error, scale factor, thermal effect, coefficients for polynomial describing the background; *U, V, W* and mixing parameters of the profile peak function, lattice constants, positional parameters and overall isotropic temperature factors. Thermal effect and profile function refinement make were fitted better than other effect in the refinement. The pseudo-Voigt description as profile shape was determined as profile set up for Rietveld refinement. Refinement profile

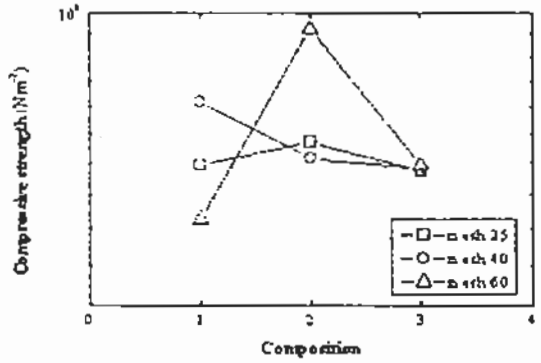

Fig. 1. The compressive strength as a function of composition.

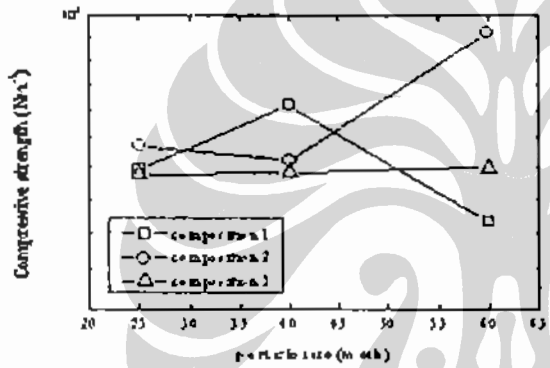

Fig. 2. The compressive strength as a function of particle size.

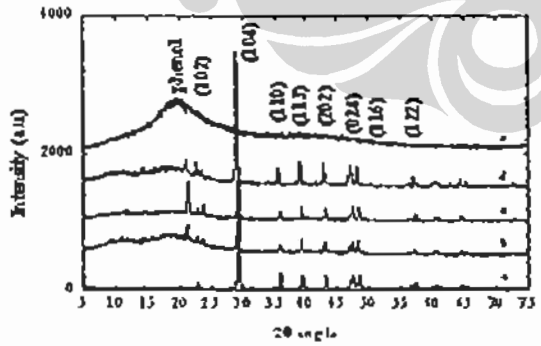

Fig. 3. The XRD spectra of marble composite, (a) marble, (b) composition 3 (67% particle of marble), (c) composition 2 (60% particle of marble), (d) composition 1 (50% particle of marble), (e) phenol.

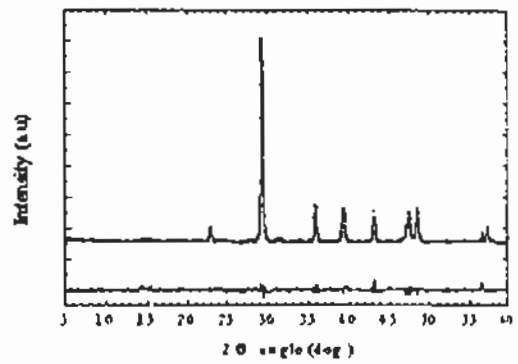

Fig.4. Rietveld plots showing the observed, calculated, difference profile, lattice constants and figures of merit for the final iteration of refinements for Citatah's marble (West Java). a = b = 4.969 Å, c = 17.026 Å.

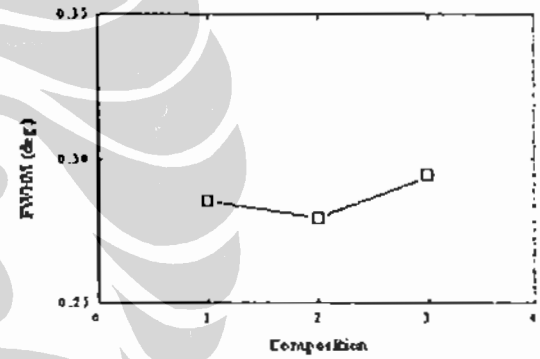

Fig. 5. The FWHM – sample for (104) plane as a function of marble composition.

function value use modern commercial Bragg-Brentano diffractometer and value of the sample broadening. Figure 4 shows Rietveld plots the observed, calculated, difference profile, lattice constants and figures of merit for the final iteration of refinements for Citatah's marble sample. The Rietveld results described the "quasi-real" structure of marble. In spite of the actual local structure is significantly different. Rietveld did constraint treatment to marble ($CaCO_3$) caused the probability of position Ca/C/O in impurity for marble was not sure.

The FWHM for (104) plane as in Figure 5. From the diffraction pattern of the composite sample with composition 1 (Fig. 5d), composition 2 (Fig. 5c) and composition 3 (Fig. 5b), the combination of 2 crystal structures that is marble crystal structure and phenol crystal structure can be seen here. Referring to the data

and the diffraction pattern, it can be concluded that composition 2 is a simple that has stronger characteristics compared with composition 1 and 3, since it has sharper diffraction pattern. The graph shows that the sample with composition 2 has FWHM smaller than that of composition 1 and 3; this indicates that the characteristics of the material is stronger and can be proved by the compressive strength value of sample with composition 2 having higher compressive strength value compared with the samples with composition 1 and 3.

Table 2. Observed and calculated X-ray powder diffraction data of $CaCO_3$.

Space Group $R\bar{3}C$ (D_{3d}^6, N° 167), rhombohedral, Z = 6, $a = b = 4.990$ Å, $c = 17.002$ Å								
$2\theta_{obs}$ (°)	d_{obs} (Å)	$(I/I_0)_{obs}$	h	k	l	$2\theta_{cal}$ (°)	d_{cal} (Å)	$\Delta 2\theta$
23.070	3.8520	29	0	1	2	23.061	3.8555	-0.009
29.455	3.0300	100	1	0	4	29.418	3.0352	-0.037
31.543	2.8340	2	0	0	6	31.550	2.8348	0.007
35.966	2.4950	7	1	1	0	35.973	2.4957	0.007
39.419	2.2840	18	1	1	3	39.419	2.2851	0
43.167	2.0940	27	2	0	2	43.165	2.0951	-0.002
47.147	1.9261	4	0	2	4	47.129	1.9277	-0.018
47.645	1.9071	17	0	1	8	47.675	1.9069	0.03
48.579	1.8726	34	1	1	6	48.532	1.8752	-0.047
56.557	1.6259	2	2	1	1	56.569	1.6263	0.012
57.401	1.6040	15	1	2	2	57.407	1.6046	0.006
58.272	1.5821	2	1	0	10	58.297	1.5822	0.025

4. CONCLUSION

Crystal structure of marble is rhombohedral and the lattice constant are $a = b = 4.969$ Å; $c = 17.026$ Å. This results of the characterization indicate that the composites and marble can be used to make tile, because they were lighter, stronger and the amount of crystals increases compared with the pure marble. This research indicates that a sample with 60% of 60 mesh marble particle can be used as a standard for composite tile. The composite sample was lighter in approximately of 40% weight reduction compared with the tile with the same volume, furthermore the cost is cheaper.

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