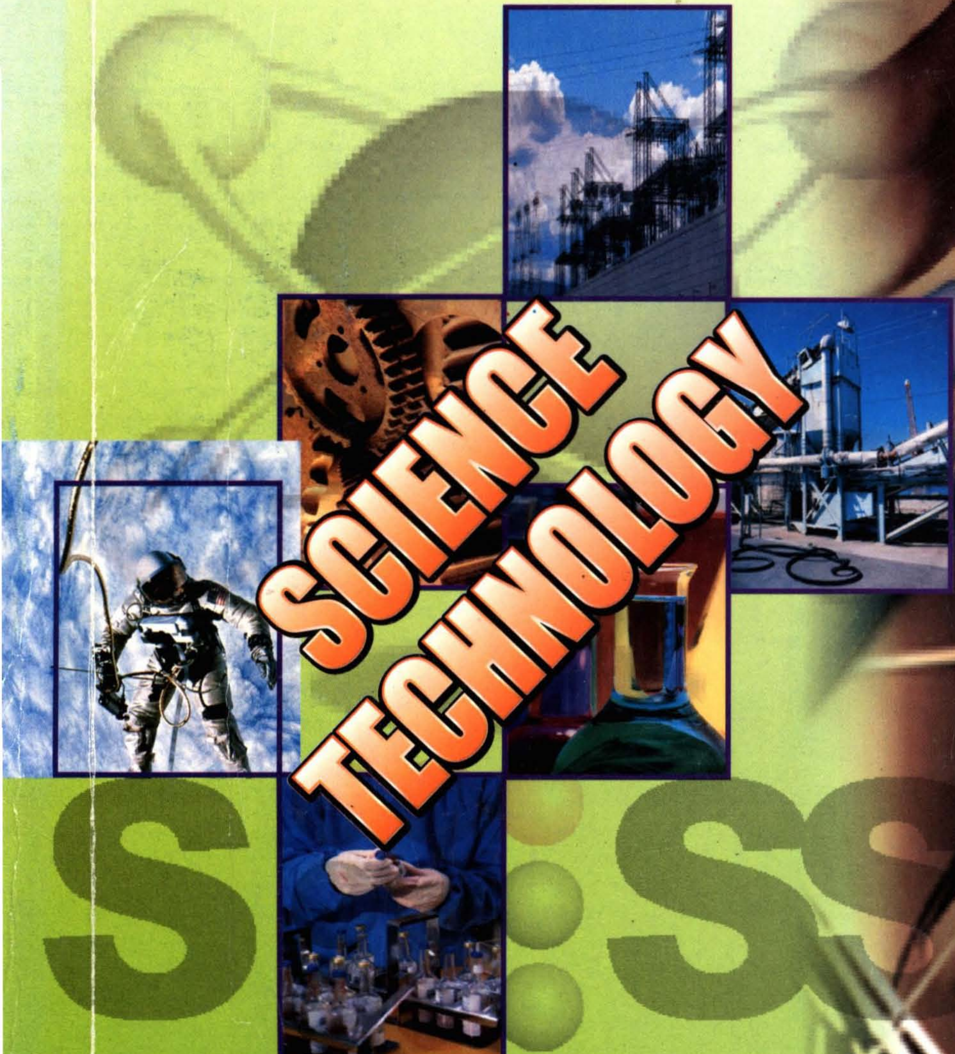
UNIVERSITI TEKNOLOGI MARA

ISO 9001:2000 No Sijil :0500132

Globalising Knowledge and Information

NATIONAL SEMINAR ON SCIENCE TECHNOLOGY & SOCIAL SCIENCES 2006



30-31 May 2006

Swiss Garden Resort & Spa

Kuantan, Pahang

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

Irzaman
S. Nurhasanah
Z. Jamal

ABSTRACT

Composite tile was made using Java's marble particles ($CaCO_3$), phenol resin and as a catalyst was used hexamethylenetetramine (HEXA), then using simple mixing method based on volume variation from 62.50 ml to 125.00 ml between matrix and hardener, based on mesh variation consisted of 25 mesh, 40 mesh 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 6.15 x 10^7 N/m^2 – 9.61 x 10^7 N/m^2, and there were two crystal structures consisted of marble crystal and phenol crystal. Crystal structure analysis of $CaCO_3$ was carried out by using the Rietveld semiquantitative analysis. The final crystal structure was rhombohedral and lattice constant were a = b = 4.989 Å, c = 17.042 Å; figures of merit were: R_p = 10.43 %; R_{wp} = 15.35 %, GOF = 3.49; R_B = 3.94 % (marble); R_B = 4.09 % (50 % of marble particle); R_B = 4.12 % (60 % of marble particle); R_B = 4.14 % (67 % of marble particle). The results of characterising indicated the composites and marble were able to use for making tile, because they were lighter, stronger and the amount of crystals increased if compared with pure marble only.

Keywords: *Composite material; physical properties; XRD. Rietveld Analysis.*

Introduction

The growth of construction industry in Java Indonesia and Italy has developed so rapidly that the research workers try to get a new alternative construction material having better quality than before which was 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 (Gibson 1994). 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 others (Pusat Pengembangan Teknologi Mineral 1987). 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 (Manggasa 1998). The waste products can be use for making composite tile as an alternative construction material.

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

The Experiment

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, then, crushing with a mortar or hammer, refining using nur mit schutzring machine. They were sieved with a 25, 40 and 60 mesh sieves to get finer particles of marble. The sample was made as shown on Table 1, and took these following steps: first, the marble particles were poured into phenol resin little by little while stirred.

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

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.

Afterwards, catalyst hexamethylenetetramine (HEXA) was added little by little and stirred until it got homogenous for 15 minutes. The matrix and the glass as the layer should be cleaned and given mirror glaze as the grease in order that the sample was not sticky to them. And then the mixture was poured onto 5 cm x 5 cm and 5 cm x 25 cm matrix and then dried in room temperature for 3 hours. At last, the mixture was removed. Finally, the sample was characterised that is, compressive strength was measured with Universal testing machine. For characterising was used 2.5 cm x 1 cm sample; crystal structure was analysed by x-ray diffraction (XRD) and Rietveld refinement. The XRD spectra were recorded on a Shimatzu type 610 diffractometer using CuK_α radiation at 30 KV and 30 mA (900 watt).

Results and Discussion

Compressive Strength Measurement

The compressive strength measurement can be completely seen in Figures 1 and 2. The figures shows that in increasing the fraction of the marble particle volume will increase the compressive strength of the composite (Gibson 1994) and has also shown that the marble particle refining will increase the value its compressive strength (Spriggs 1996). The compressive strength value of M260 (composition 2, 60 mesh) is 9.61×10^7 N/m^2, this value is lighter than 8.00×10^7 N/m^2 as the compressive strength value of a tile (SNI 13-0089-1987. 1987). 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.

Crystal Structure Measurement

Figure 3 shows XRD spectra of Java's marble composite various compositions. 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 (Delgado et al. 2003; Rietveld 1969; Raksa et al. 2004) was carry out using the program (Li et al. 1991; Hill 1986). The structure of marble in Citatah West Java Indonesia is formed by calcium carbonate ($CaCO_3$) (Irzaman et al 1999), space group R3C (Hahn, T. 1983), rhombohedral crystal system found in the ICDD (JCPDS, 1997) was used as the initial model of $CaCO_3$.

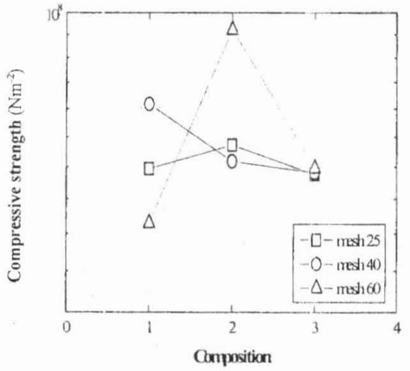


Fig. 1: The compressive strength as a function of composition

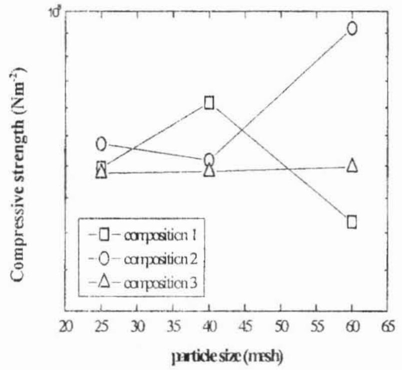


Fig. 2 The Compressive strength as a function of particle size

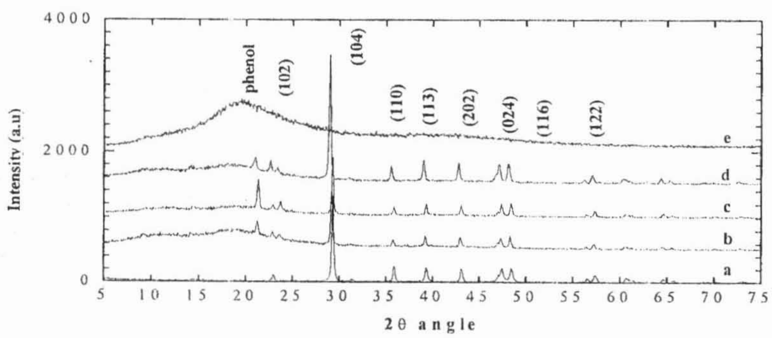


Fig. 3: The XRD spectra of Java's marble composite, (a) marble, (b) composition 3 (67% particle of marble), (c) composition 2 (60% particle of marble), (d) composition 1 (50% particle of marble), (e) phenol

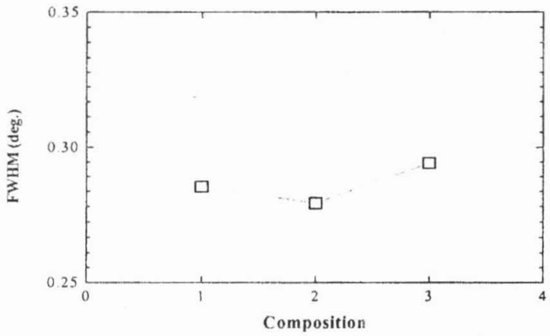


Fig. 4: The FWHM – sample for (104) plane from Rietveld refinement as a function of Java's marble composition.

The final refinement involved 21 parameters and 10 iterations, including zero shift, one scale factor, three asymmetry parameter, six coefficients for polynomial describing the background; *U, V, W* and mixing parameters of the pseudo-Voigt peak-shape function, unit cell parameters, positional parameters and overall isotropic temperature factors for each phase. The final lattice constant were $a = b = 4.989$ Å; $c = 17.042$ Å (in literature a = b = 4.990 Å; c= 17.002 Å [12]); figures of merit were : $R_p = 10.43$ %; $R_{wp} = 15.35$ %, GOF = 3.49; $R_B = 3.94$ % (marble); $R_B = 4.09$ % (50 % of marble particle); $R_B = 4.12$ % (60 % of marble particle); $R_B = 4.14$ % (67 % of marble particle); and FWHM for (104) plane as in Figure 4. From the diffraction pattern of the composite sample with composition 1 (Fig. 3d), composition 2 (Fig. 3c) and composition 3 (Fig. 3b), 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.

Conclusion

This research indicated that is a sample with 60 % of 60 mesh marble particle can be used as the standard of the material for composite tile. Characteristic of this sample is lighter, stronger, more crystalline and cheaper. Composite sample was made lighter with approximately 40 % weight reduction compared with the tile with the same volume. In addition, it costs is cheaper because it is made using phenol which is cheapest thermoset resin and the waste of marble tile production.

Acknowledgements

This work was supported by RUT XII Project, KMNRT and LIPI, The Republic of Indonesia under contract No. 55/SK/RUT/2005 and Short Term Research Grant from Northern Malaysia University College of Engineering, Malaysia under contract No. KUKUM/R&D/9003-00030(1)/2005.

References

Delgado, G.E., Mora, A.J., Betancourt L. & Sagredo, V. (2003). *Phys. Stat. Sol (a)*, 199 (3): pp. 373.

Gibson, R.F. (1994). *Principle of Composite Material Mechanics*. New York. USA: McGraw Hill, Inc.

Irzaman, Y. Sudiana & M. Hikam. (1999). *Proc. Third Workshop on Electro Communication and Information*. ITB

Bandung-Indonesiap: pp. 4 - 40.

JCPDS. (1997). *Powder Diffraction File (Set 1 – 5)*. International Centre for Diffraction Data. CD- Room.

Li, J.Y., Li, D.Y. & O'Connor, B.H. (1991). *Program PCRTVW-PC Version of Rietveld Program LHPM 10 and Associated Program Weigth*. Australian X-ray Analytical Association: pp. 19.

Manggasa, A. (1998). *Pengembangan Pembuatan Ubin Marmer dengan Memanfaatkan Limbah Pengolahan Marmer*. Laporan Penelitian. Balai Industri Ujung Pandang. Badan Penelitian dan Pengembangan Industri dan Perdagangan. Departemen Perindustrian dan Perdagangan. (*In Indonesia*).

Pusat Pengembangan Teknologi Mineral. (1987). *Bahan Galian Industri, Batu Pualam (marmer)*. Departemen Pertambangan dan Energi. (*In Indonesia*).

R.J. Hill & C.J. Howard. (1986). *A Computer Program for Rietveld Analysis of Fixed Wavelength X-ray and Neutron Powder Diffraction Patterns*. New South Wales: Australian Atomic Energy Commission Research Establishment

Raksa, Y.T., Hikam, M. Irzaman, Darmasetiawan, H. (2004). *Rietveld Analysis of Ferroelectric $PbZr_{0.525}Ti_{0.475}O_3$ Thin Films*. Ceramics International, 30: pp. 1483.

Rietveld, H.M. (1969). Crystallogr. *J. Appl*, **2**: pp. 65.

SNI 13-0089-1987. (1987). *Mutu dan Cara Uji Marmer*. Dewan Standardisasi Nasional. (*In Indonesia*).

Spriggs, R.M. (1996). *Grain Size Effect in Polycrystalline Ceramics on Materials Science Research*. New York: Plenum Press.

T. Hahn. (1983). *International Tables for Crystallgr. A: Space Group Symmetry*. Boston: D. Reidel Publishing Company: pp. 540.

IRZAMAN & S. NURHASANAH, Laboratory of Material Physics, Department of Physics, FMIPA IPB, Kampus IPB Darmaga Gedung Wing S Bogor, Indonesia – 16680. irzaman@kukum.edu.my, irzaman@yahoo.com.

Z. JAMAL, School of Microelectronic Engineering Northern Malaysia University College of Engineering.