STM JOURNALS

Journal of Polymer & Composites
ISSN: 2321-2810 (Online), ISSN: 2321-8525 (Print)
Volume 8, Issue 2
www.stmjournals.com

Development of Refractory Synthesized from Waste Ceramic Fiber and Chamotte

Do Quang Minh[1,]*, Nguyen Vu Uyen Nhi[2], Nguyen Hoc Thang[3,*]
[1]Associate Professor, Faculty of Materials Technology, Ho Chi Minh City University of Technology (HCMUT) – Vietnam National University (VNU), Ho Chi Minh City, Viet Nam
[2]Research Scholar and Lecturer, Faculty of Materials Technology, Ho Chi Minh City University of Technology (HCMUT) – Vietnam National University (VNU), Ho Chi Minh City, Viet Nam
[3]PhD. Eng., Faculty of Chemical Technology, Ho Chi Minh City University of Food Industry, Ho Chi Minh City, Viet Nam

Abstract
Research on refractory materials is always essential in thermal engineering to ensure both heat savings and environmental protection. Raw materials used initially have a great influence on the production process and product properties. In addition, the refractory products are tested under conditions of high temperature and varying intensity, such as tests of thermal shock resistance. In this study, new refractory composites were prepared by mixing waste ceramic fibers with chamotte materials and heated at 1220^{O}C for 1 hour. The refractory composites have low values of the volumetric weight from 0.87 to 1.32 g/cm^3 which are belong to group oflightweight materials. More important, the strength values of the products are high with testing of bending and compressive strength. Experimental results also showed that the thermal shock resistance of these synthesized materials has high quality in comparison with others referential chamotte refractory materials. Moreover, the coefficient of thermal expansion of the composite is lower than the same types of materials. Analysis of scanning electron microscope image (SEM) shows that the ceramic fiber distribution in the material has the effect of preventing the development of cracks. The material has been applied as resistors, hot gas pipe connectors, industrial furnace repair mortar.

Keywords: *Ceramic fiber,Chamotte,Refractory, Thermal expansion, Thermal shock resistance*

****Author for Correspondence:*** E-mail: mnh_doquang@hcmut.edu.vn and thangnh@hufi.edu.vn

INTRODUCTION

Refractory materials are types of heat resistant material used as linings for furnaces, high-temperature reactors and others. In addition to being resistant to thermal shocks and physical phenomena caused by heat, refractory materials are also resistant to physical wear and corrosion from chemicals. Refractory materials are more heat-resistant than iron and are required to be applied at high temperatures where other materials are not able to withstand this harsh heat environment [1-3].

Refractory consists of types of materials which have the characteristics of high thermal shock resistance as mentioned above, according to different temperatures, for various time periods, and under different applied conditions. There are many components of refractories manufactured in many different shapes and forms to suit various types of applications. A common feature of refractory materials will be subjected to high temperatures while being repaired. Refractory products are divided into two types: bricks or fired forms and amorphous refractories or specialized materials. Refractory linings are produced from bricks and prefabricated concrete or from specialized refractory materials such as resins, refractory cements, sprayed or compacted mixtures, or a combination of both [4, 5].

Many refractory products have similar shapes to traditional building bricks. However, there are many different shapes and styles such as some flame retardant materials are smaller and maybe have complex and sophisticated shapes;

others are large and can weigh tons in the form of prefabricated blocks [2, 5-8].

Fig. 1: *High Alumina Refractory Brick.*

Fig. 2: *Porous Insulation Bricks.*

Refractories are materials capable of retaining its properties at high temperature. They have a non-metallic main components with combinations of minerals known as alumina, fireclays, bauxite, chromite, dolomite, magnesite, silicon carbide, zirconia, and others. Refractories must have responses the basic requirements: high thermal shock resistance, inertness in chemical reactions and coefficient of thermal expansion within the permissible range of high temperatures [9-11].

Conventional refractories are classified into three main groups, which are acidic refractories, basic refractories, and neutral refractories.

Chamotte refractory materials are manufactured from chamotte stone with refractory clay and fired at high temperatures of 1450^{o}C. Tests of thermal shock resistance of the chamotte refractory showed that it has around 30 times before destroyed. The processes are carried out at 950^{o}C for 1 hour and then put in cold water.

Ceramic wool is a kind of product made of ceramic fiber, in rolls, able to insulate at high temperatures from 1260^{o}C to 1430^{o}C. Ceramic fiber has special uses for kilns, ceramic furnaces. Steel furnaces, kilns of ceramic tiles, food ovens, fireproof windows. Ceramic wool is manufactured to serve thermal insulation projects at high temperature or exposure to fire up to 1600^{o}C.

Ceramic fibres are low thermal absorption and conductivity, more flexible and high compressive strength, accurate size with high flatness, homogeneous structure, easy to apply and repair, high chemical resistance and heat resistance [11-14].

Ceramic fibers are small sized fibers made from oxides such as Al_2O_3, SiO_2, and ZrO_2, with high thermal shock resistance [12]. Therefore, the combination of waste ceramic fibers and chamotte refractory aims to improve the thermal endurance of these materials [3,4]. Moreover, this is also the solution for environmental treatment caused by the waste ceramic fibers.

MATERIALS AND METHODOLOGY

Raw materials are Trai Mat chamotte (Lam Dong province, Viet Nam), Tan Uyen clay (Binh Duong province, Viet Nam), ceramic fiber from Isowool company, and Al(OH)3 from Tan Rai bauxite factory (Lam Dong province, Viet Nam). Raw meals were mixed from chamotte with particle sizes around 1-2mm, ceramic fiber, clay, $Al(OH)_3$, and 30% water. The various ratios among raw materials are shown in Table 1.

Table 1: *Ratios of the Raw Materials in the Experimental Mixtures.*

Raw materials	Sample					
	S_0	S_5	S_{10}	S_{15}	S_{20}	S_{25}
Tan Uyen clay	18	18	18	18	18	18
Chamotte	80	75	70	65	60	55
$Al(OH)_3$	2	2	2	2	2	2
Ceramic Fiber	0	5	10	15	20	25

The mixtures were then formed in the cylindrical samples with a diameter of 80 mm and height of 100 mm. The samples were heated at 1220^{o}C for 1 hour to refractory composite products. Finally, the refractory specimens were tested engineering and thermal properties. In which, the thermal shock resistance was carried out by the number of times the sample heated at 950^{o}C and cooled in cold water without broken.

Samples with the best thermal shock resistance were selected for characteristics of microstructure and thermal properties using SEM (scanning electron microscopy), thermal analysis (DTA-TG), and coefficient of thermal expansion (DIL-dilatometer). Evaluation of characteristics among the samples was carried out to have conclusions about the role of ceramic fibers in the refractory composite materials.

Thermal stress δ in a material is usually calculated by the formula [5]:

$\delta = E.\alpha.\Delta T$(1)

In which,

E - modulus of elasticity

α - coefficient of thermal expansion (1/^{o}C)

ΔT - Temperature difference (^{o}C)

The above formula shows that the thermal stress will be decreased in case of material haslow the coefficient of thermal expansion. It means that refractory composites must be increased the values of thermal shock resistance.

RESULTS AND DISCUSSION

In this section experimental results are discussed and analyzed. The section describes properties of raw materials, engineering properties of the refractory composites and coefficient of thermal expansion and several refractory samples for application.

Properties of Raw Materials

For isowool ceramic fiber1400, it has diameter at 2.8 µm, a length of 250 µm, and specific gravity of 2.8 g/cm^3. Trai Mat kaolin was characterized for thermal analysis (DTA-TG) from room temperature to 1220^{o}C to obtain Trai Mat chamotte as shown in Figure 3.

Figure 3 shows that total mass change of the kaolin sample is around 10.8% including values of water content and loss on ignition. The mass loss is related to physico-chemical reactions of clay minerals at different temperatures known as follows:

From room temperature to 200^{o}C: water vaporization of the kaolin sample;

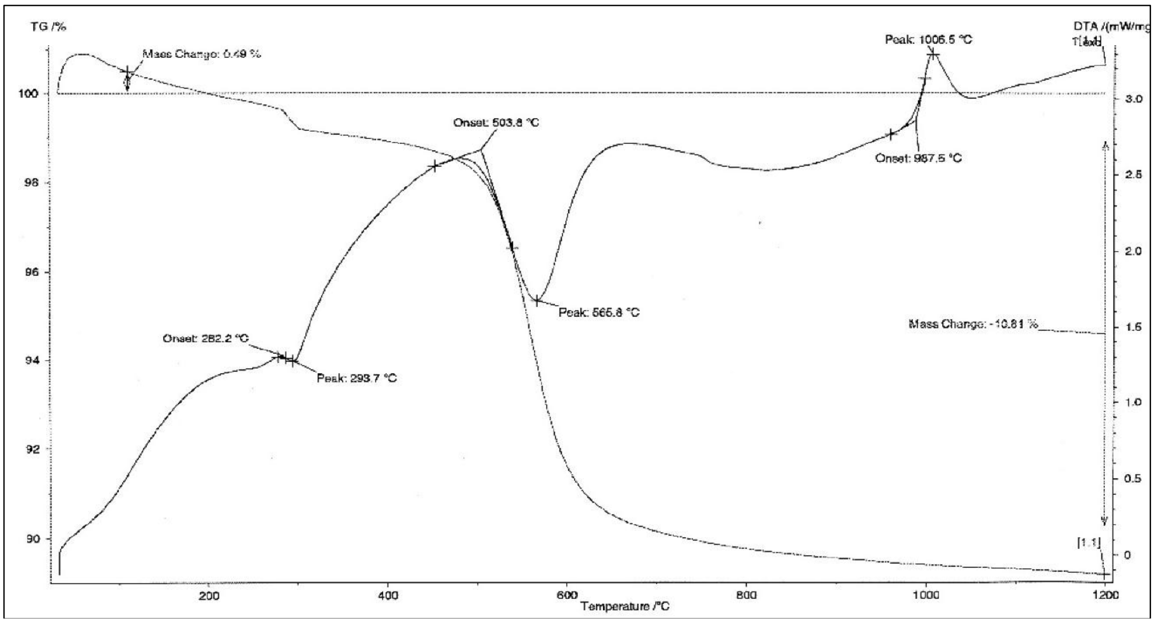

Fig. 3: *Physico-chemical Reactions of Trai Mat kaolin to form Chamotte Sand using DTA-TG.*

From 200°C to 400°C: remaining water vaporization and decomposition of organic matter, especially at 282°C and 293°C;

At 565°C, the kaolin sample has endothermic reactions with dehydroxylization of clay minerals in kaolin known as kaolinite change into metakaolinite ($Al_2O_3.2SiO_2$) in the following reaction:

$$A1_2O_3.2SiO_2.2H_2O \rightarrow Al_2O_3.2SiO_2 + 2H_2O$$

At 1006°C, the crystalline products were formed from this amorphous intermediate phase with a prominent exothermic reaction. In this case mullite ($3Al_2O_3.2SiO_2$) crystals appeared with phase transition reaction from metakaolinite:

$$3(Al_2O_3.2SiO_2) \rightarrow 3Al_2O_3.2SiO_2 + 4SiO_2$$

The crystals of mullite ($3Al_2O_3.2SiO_2$) and quartz (SiO_2) were detected bythe XRD pattern of Trai Mat chamotte sand sample which heated at 1220°C for 1 hour (Figure 4)

The raw materials (Tan Uyen clay, Trai Mat chamotte, Isowool ceramic fiber) were tested for chemical composition using a method of x ray fluorescence (XRF). The results in Table 2 show that the raw materials contain high silica and aluminum oxides from 49.7 to 64.1% and 22.9 to 44.1%, respectively. In addition, isowool ceramic fiber also has 15.0% ZrO_2,Trai Mat chamotte has no value of loss on ignition (L.O.I) while Tan Uyen clay contains high value of L.O.I at 6.7%.

***Table 2:** Chemical Compositions of Raw Materials (% wt.)*

Oxides	Tan Uyen clay	Trai Mat chamotte	Isowool ceramic fiber1400
SiO_2	64.1	52.7	49.7
Al_2O_3	22.9	44.1	35.0
Fe_2O_3	0.9	1.5	0
K_2O	2.8	1.1	-
ZrO_2	0	0	15.0
L.O.I	6.7	0	0

Engineering Properties of The Refractory Composites

The refractory samples were prepared with ratios of raw materials as mentioned in Table 1 and heated at 1220°C for 1 hour. The specimens then were tested for engineering properties such as volumetric weight (γ, g/cm^3), compressive strength (R_{CS}, MPa), bending strength (R_{BS}, MPa) and thermal shock resistance (times). The experimental results are shown in Tables 3 and 4.

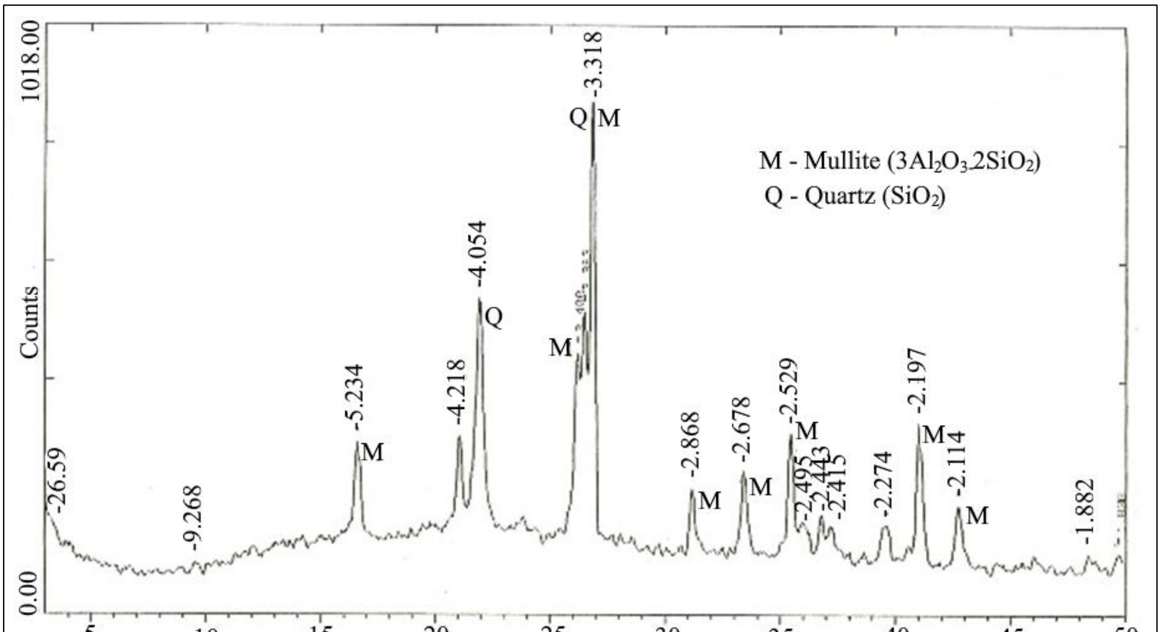

***Fig. 4:** Crystals of Mullite(M) and Quartz (Q) in Phase Compositions of Trai Mat Chamottesand after Fired at 1220°C for 1 Hour.*

Table 3:*Volumetric Weight and Thermal Shock Resistance of the Refractory Composites.*

Samples	Volumetric weight,γ (g/cm^3)	Thermal shock resistance (Times)
S_0	0.87	>30
S_5	1.32	>30
S_{10}	1.06	50
S_{15}	1.06	50
S_{20}	1.03	50
S_{25}	1.00	50

Table 4: *The Strength Values of the Refractory Composites.*

Samples	Bending strength R_{BS} (MPa)	Compressive strength R_{CS} (MPa)
S_0	3.09	27.24
S_5	7.10	54.61
S_{10}	36.06	96.58
S_{15}	44.58	145.11
S_{20}	32.75	72.24
S_{25}	30.26	63.66

For volumetric weight, the refractory samples have low values from 0.87 to 1.32 g/cm^3. In which, the S_5 sample is the heaviest material at 1.32 g/cm^3 and others samples are around 1.00 g/cm^3. For thermal shock resistance, the refractory composites have high values upto 50 time except for samples S_0 and S_5 less than 30 times because of containing low isowool ceramic fiber1400. It is noted that the samples with over 10% isowool ceramic fiber1400 have the values of thermal shock resistance more than 50 times.

For strength, the refractory specimens have bending strength from 3.09 to 44.58 MPa with the lowest values of the sample S_0 contained 0% isowool ceramic fiber. The sample S_{15} with 15% isowool ceramic fiber has the highest values of bending strength and compressive strength of 44.58 and 145.11 MPa, respectively. The refractory specimens have a compressive strength from 27.24 to 145.11 MPa. Development of the strength in the refractory samples related to the sintering process of ceramics at high temperature. The samples with high isowool ceramic fiber (S_{20} and S_{25}) were decreased the strength values because ceramic fibers with a high content inhibited the diffusion and sintering reaction of the solid powders in the mixtures.

Figures 6 and 7 are microstructure of the refractory samplesS_o and $S_{15}$with ceramic fibers rodsand solid phases. The structure of the sample S_0 is background of solid phase particles while the sample S_{15} contains the ceramic fiberrods bonded to surrounding solid phase particles. The rod structures inhibited the development of micro-cracks in the refractory chamotte materials, which improving the mechanical and thermal properties of the sample S_{15}.

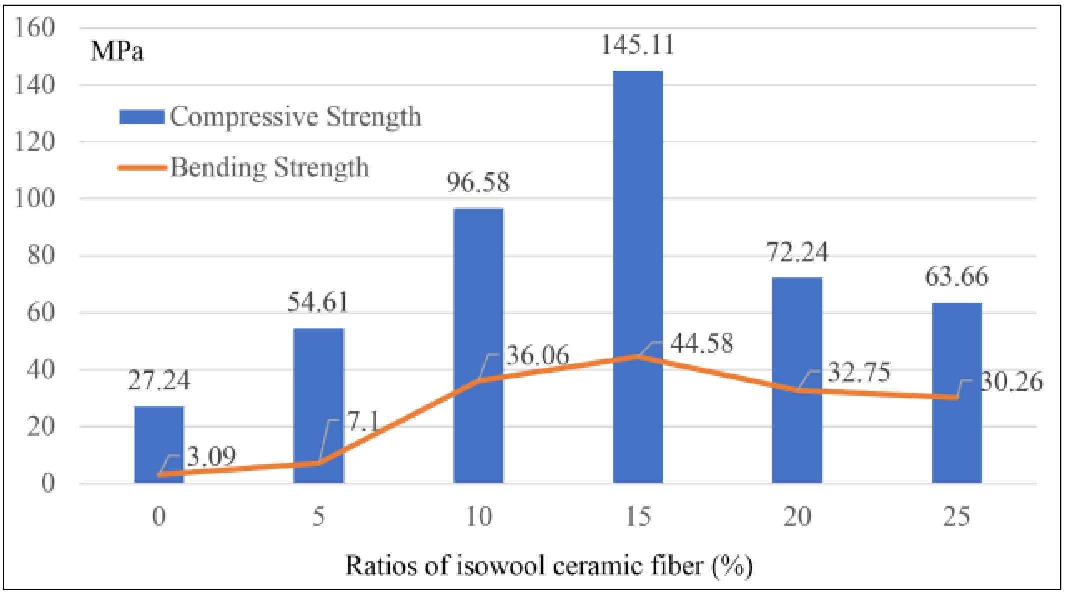

Fig. 5: *Compressive and Bending Strength (MPa) of the Refractory Composite Materials Produced from Chamotte Sand and Ceramic Fiber.*

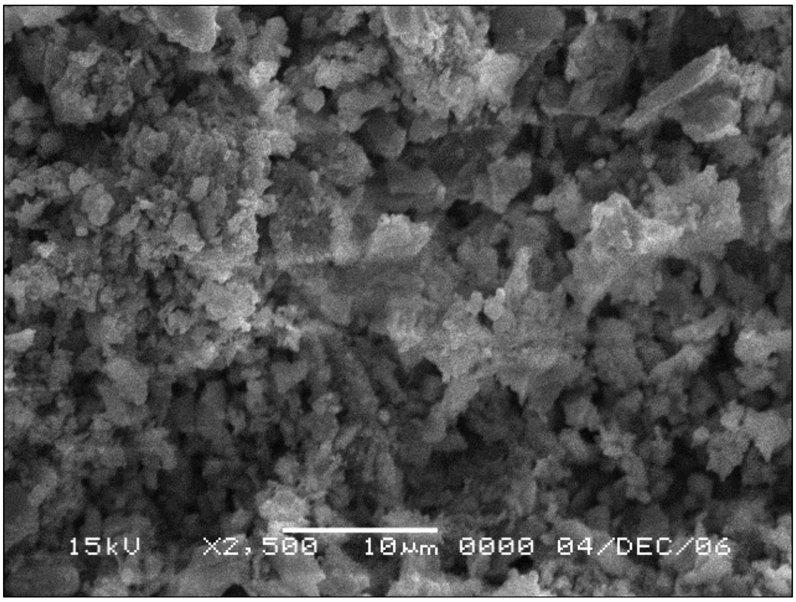

Fig. 6: *Microstructure of the Refractory Samples S_{0}.*

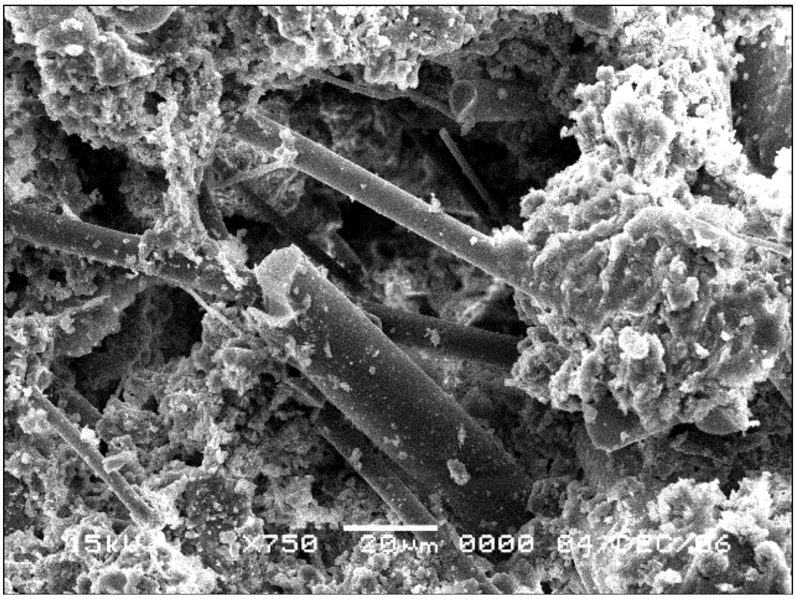

Fig. 7: *Microstructure of the Refractory Samples S_{15}.*

Coefficient of Thermal Expansion and Several Refractory Samples for Application

The refractory samples of S_0 and S_{15} were also characterized for coefficient of thermal expansion using Dilatometerwith the results as shown in Figures 8 and 9.

The coefficient of thermal expansion in Figures 8 and 9 are determined by the linear equation:

$$y = ax + b \quad (1)$$

In this equation,

$y = \Delta L/L_0$ is length decrease of the sample compared to the original length L_o,

*x*is temperature (^oC),

a and *b* are experimental coefficients.

Both the refractory samples of S_0 and S_{15} have the coefficient of thermal expansion with linear lines divided into 3 parts related to thermal changes of phases in the refractory composites. It was based on the values from Figures 8 and 9, the linear equations (y) are determined in the ranges of the various temperature as follows:

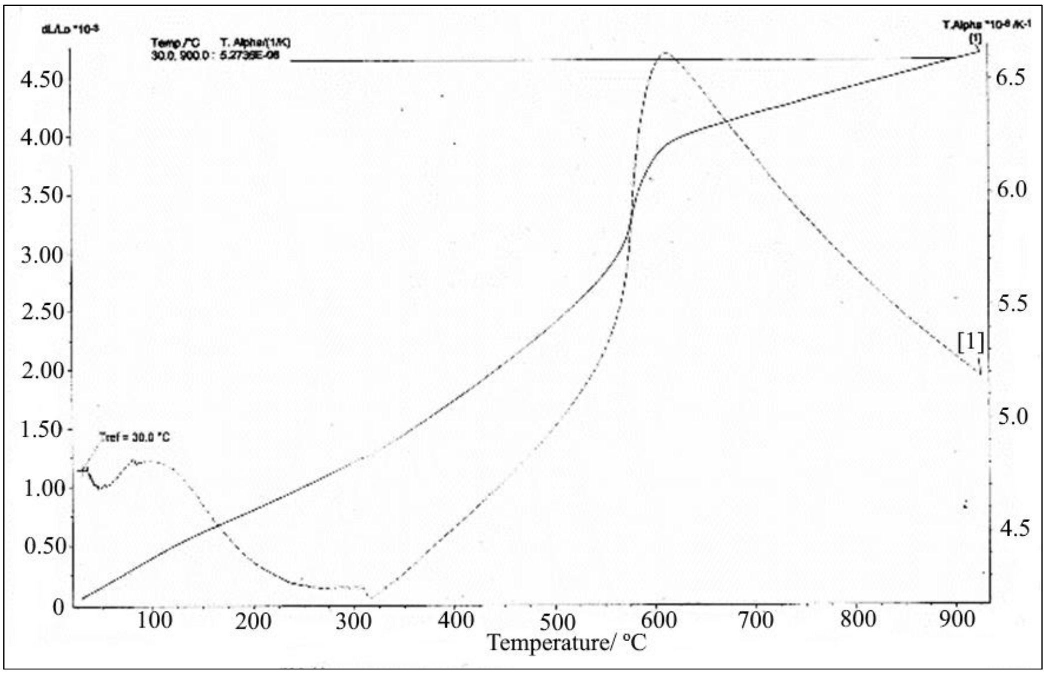

Fig. 8: *The Coefficientof Thermal Expansion of the Refractory Composite without Isowool Ceramic Fiber (sample S_0).*

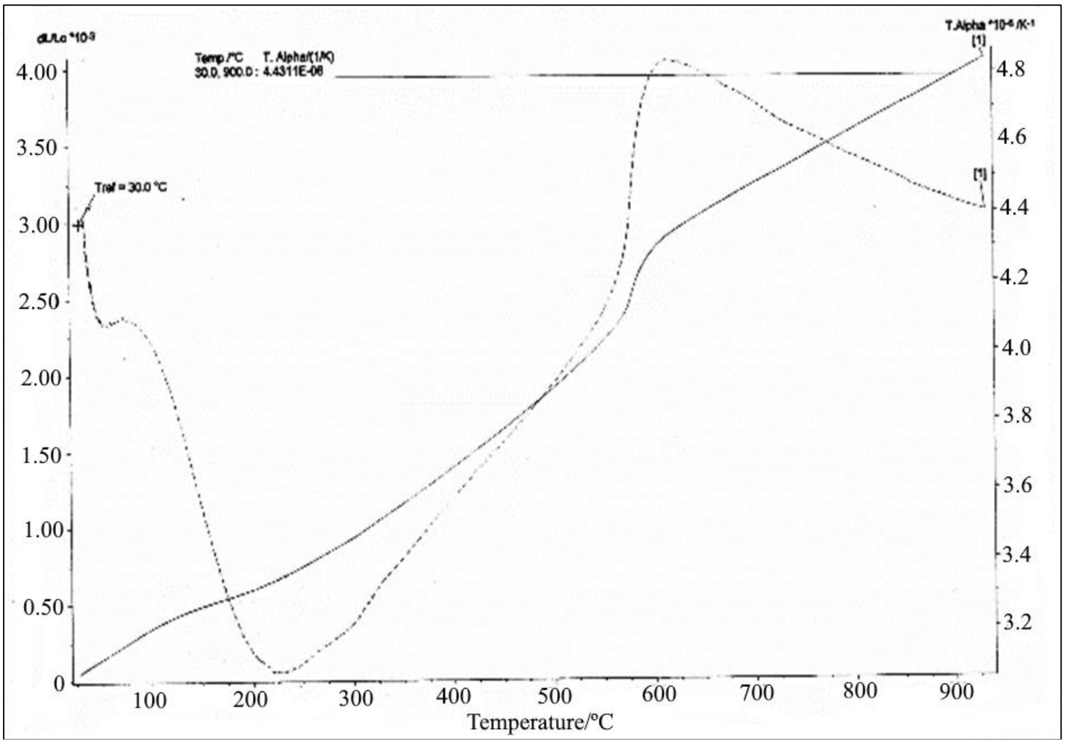

Fig. 9: *The Coefficientof Thermal Expansion of the Refractory Composite with 15% Isowool Ceramic Fiber (sample S_{15}).*

From room temperature to 560^{O}C, the equations for coefficients of thermal expansion of the samples

S_0:*y=[0.00437.x+2.83076].10^{-6} (1/ ^{o}C)*

S_{15}:*y= [0.00334.x+2.31466].10^{-6}(1/ ^{o}C)*

From560^{O}C to 610^{O}C, both of the samples have phase structural transform of the quartz crystals (SiO_2) in the refractory composites:

β-quartz↔-quartz

From 610^{O}C to 950^{O}C, the equations for coefficients of thermal expansion of the samples

S_0:*y=[0.00469.x + 9.42050].10^{-6} (1/^{O}C)*

S_{15}:*y= y=[0.00140.x + 5.68986].10^{-6}(1/^{O}C)*

The coefficient of thermal expansion of the sample S_{15} is lower than that of the sampleS$_o$.This leads tothe thermal stress δofsample S_{15}islower and thatis also the reason which the sample S_{15} hashigh thermal shock resistance than the sample S_o.

The investigation was carried out production of several refractory samples applied in furnace as shown in Figure 10.

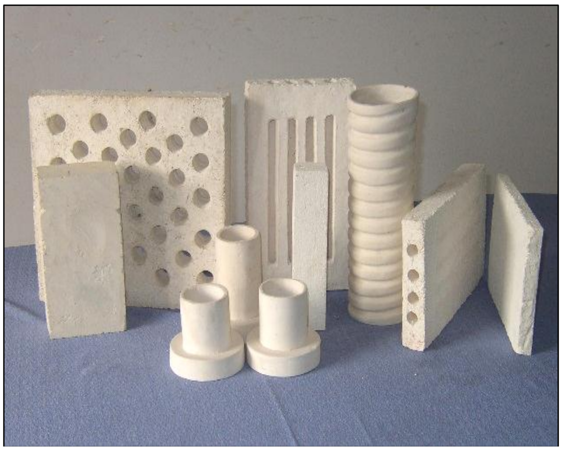

Fig. 10: *Some Practical Applications of the Refractory Composites from Waste Ceramic Fiber and Chamotte Material.*

CONCLUSION

Refractory composite materials are applied in many fields with the different production technology. This study used isowool ceramic fibers, Tan Uyen clay, and Trai Mat chamotte sand for producing the refractory. These refractory composites are fired at a relatively low temperature (1220^{o}C), but its quality is still guaranteed for thermal shock resistance. The ceramic fibers created bonding networks in the chamotte background to increase the thermal shock resistance and the strength.Besides, the fiber rods prevented the development of microcracks in the material. Therefore, the waste ceramic fiber should be used as a raw material to producerefractorymaterials. This will contribute to reduce the emissions of ceramic fibers causing environmental pollution.

Nomenclature

δ	:	Thermal Stress
E	:	Modulus of Elasticity
α	:	Coefficient of Thermal Expansion
ΔT	:	Temperature Variation
DTA	:	Differential Thermal Analysis
XRD	:	X-Ray Diffraction
L.O.I	:	Loss On Ignition
SEM	:	Scanning Electron Microscopy

REFERENCES

1. EijiHorie, Ceramic fibre insulation theory and practice-The energy conservation center, Tokyo,1997.
2. Černý, R., Poděbradská, J., Toman, J., Jiříčková, M., Mňahončáková, E., Rovnaníková, P., Bayer, P., High Temperature Properties of Fibre Reinforced Cement Composites, *Proceedings of the International Conference on Application of Codes, Design and Regulations*, 2005, 403-412.
3. Holčapek, O., Reiterman, P., Konvalinka, P., Fracture Characteristics of Refractory Composites Containing Metakaolin and Ceramic Fibers, *Advances in Mechanical Engineering*, 2015, 7(3), 1-13.
4. Ondřej, H., Jaroslava, K., Pavel, R., Fiber Reinforced Concrete with Application in Civil Engineering,*Advances in Civil Engineering*, 2018, 2018,1-11.
5. Kingery, W.D., Bowen,H.K., Uhlmann, D.R.,*Introduction to Ceramic*, 2nd edition, Wiley, 1976.
6. Groover, P. Mikell, *Fundamentals of Modern Manufacturing: Materials, Processes, and Systems*, John Wiley & Sons, 2010.
7. Deer, W.A., Howie, R.A., Zussman, J., *An introduction to the rock-forming minerals*, 2nd edition, Harlow: Longman, 1992.
8. Káosek-Wawrzyn, E., Maáolepszy, J., Murzyn, P., Sintering Behavior of Kaolin with Calcite, *Procedia Engineering*, 2013, 57, 572 – 582.
9. Mohtor, N.H., Othman, M.H.D., Ismail, A.F., Rahman, M.A., Jaafar, J., Hashim, N.A., Investigation on the effect

of sintering temperature on kaolin hollow fibre membrane for dye filtration,*Environmentalscienceand pollutionresearch*, 2017, 24, 15905–15917.

10. Do, Q.M., Nguyen, T.T.L., Pham, V.T.H.Q., Nguyen, H.T., The Novel Crystalline Glaze for Decoration of Ceramic Pottery, *Materials science forum*, 2020, 987, 165-170.
11. Ravichandran, G.,Halloysite nanotubes-A novel reinforcing material for future applications, *Journal of polymer and composites*, 2019, 7(1), 34-40.
12. Patnaik, S.C., Maharana,S., Adak, S., Mishra, A.,A Study on the Sintering Behavior of Al_2O_3/Y_2O_3 Doped ZrO_2 Composites, *Journal of polymer and composites*, 2014, 2(1), 1-8.
13. Nguyen, H.T., Gallardo, S.M., Bacani, F.T., Hirofumi Hinode, Do, .M., Do, M.H., Promentilla, M.A.B., Evaluating thermal properties of geopolymer produced from red mud, rice husk ash and diatomaceous earth, *ASEAN engineering journal*, 2015, 4(1) 51-65.
14. Nguyen, H.T., Pham, T.K., Abdullah, M.M.A.B., Lightweight heat resistant geopolymer-based materials synthesized from red mud and rice husk ash using sodium silicate solution as alkaline activator, *MATEC Web of Conferences*, 2017, 97, 01119.

Cite this Article

Do Quang Minh, Nguyen Vu Uyen Nhi, Nguyen Hoc Thang. Development of Refractory Synthesized from Waste Ceramic Fiber and Chamotte. *Journal of Polymer & Composites*. 2020; 8(2): 101–109p.