

Thermophysical characterization of composite clay Materials doped by the copper powder according to the Temperature

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Abstract: - The objective of this work is the development of a composite material of clay doped with copper powder and with thermal properties likely to give it a great importance in the ceramic industry, has been the subject of increased attention in recent decades because of their multiple uses as thermal insulators in the fields of ceramics and heterogeneous catalysis. Indeed, this study is based on a global approach of the properties of the material, accompanied by a thorough analysis of the thermophysical characteristics. The thermal conductivity of this material was determined as a function of copper content and temperature. Experiments have shown that thermal conductivity increases with the copper content and decreases with temperature, the experience made by an apparatus called CT-METRE. A theoretical model of the composite material allowing to approach more finely the heat transfers and also to validate the experimental results obtained.

Key-Words: - clay, clay content, copper content, thermal conductivity.

Received: October 16, 2019. Revised: May 3, 2020. Accepted: May 9, 2020. Published: May 19, 2020.

1 Introduction

As for composite materials they are commonly defined as being reassembly of two or more materials of complementary properties. Generally composite materials consist of two essential elements, whose contributions are complementary, the matrix and the reinforcement. The matrices are presented, most often as lightweight materials and feeble mechanical resistance. Their essential role in composites is to maintain the cohesion of the collection [1][2]. By reinforcement, we mean any element probable to improve the properties of the matrix, in which it is incorporated, its presence in a composite material improves its mechanical and thermal performance [3] [4]. The material studied in this work is obtained by mixing copper and clay into powders with water for petrification which results in a solid solution of substitution of thermo-physical properties depending on its composition and temperature. These copper powders make it possible to improve the thermal performance of clay by modifying its thermo-physical characteristics, which are the thermal conductivity obtained

experimentally by a "CT-METRE" apparatus and also by a theoretical model. In our work consists in studying by experiment the relation between this thermal quantity and the copper content as well as their variation with the temperature, therefore the study of the influence of the addition of copper powder in different contents following 5 %, 10%, 15%, 20% and 25%, on the thermal behavior of the clay with respect to its composition and temperature [5] [6][7]. After the calculations and the experiment we are led to conclude that the conductivity increases with the copper content and decreases with the temperature, which is due to the adsorption of electropositive atoms of copper on sites with high negative charge of clay particles as well as increased transport of heat by copper electrons [8].

2 Material development method

The procedure for crushing clay and copper in a crusher: in order to obtain ultrafine powders of copper and clay, sieve is then sieved with a sieve (diameter 1 μm), then in prepares five pairs of samples (the corresponding copper-doped agile) of

the following different percentages: 5%, 10%, 15%, 20%, and 25%. After preparing the five pairs of samples the homogeneses with water to obtain them in pasty form, then they are placed in a mold of dimensions (7cm * 7cm * 5cm) [9]. To develop the material in simple rectangular parallelepiped geometric form, the heat treatment is carried out in the oven (Fig. 2) at 40 ° C for 72 hours in order to reduce the water content and finally or compacts the material to minimize its porosity [10] [11].

2.1 Presentation of the assembly

The measurement of the thermal conductivity of the material is carried out by conductimetry using an appliance <<CT-METRE>>, (Fig.1) equipped with accessories like specific sonde for solid materials, a glove box consisting of a thermocouple which serves to fix the temperature and an interface that couples the editing with a computer to display and process the measured numerical values.

The CT-METRE, an easily transportable device, was developed with the aim of making it possible to accurately assess the thermal characteristics of a certain number of materials, such as: brick, rocks, earth, aerated concrete, bitumen, powdered substances, resins or complex products, etc.

The operating principle of consists, thanks to the association of a heating element and a temperature capture (both associated in the same sonde), to measure the rise in temperature undergone by the capture, during a heating period chosen by the user depending on the material to be tested and the type of probe used (the ring sonde or the wire sonde).

2.2 Procedure

The CT-METRE consists of two elements:

- a) The control unit, responsible for generating the heating power and interpreting the temperature rise curve induced in the material to be tested.
- b) The sonde responsible for transmitting the heating power and collecting the induced temperature.

First connect the ring sonde (Fig. 3) with the plug connected to the appliance <<CT-metre>> (Fig.1), then it is inserted between two samples of the same size and same shape so that it sweeps the sample in volume, the operator starts the measurement by pressing the MEASURE button. From this moment, the cycle proceeds automatically and at the end of the programmed measurement time, the display transmits the expected results. This appliance

measures the thermal conductivity of materials in steady-state.



Fig. 1. General diagram of the <<CT-METRE>> allowing the measurement of the thermal conductivity of the samples

These four samples have been studied in rectangular forms, each sample is divided in two, the heat treatment is carried out in the oven at 40 ° C for 72 hours in order to reduce the water content and finally or compacts the material to minimize its porosity. (Fig. 2)



Fig. 2. The samples in the oven

The ring sonde is made up of a flexible printed circuit (thickness 0.2 mm-dimension 60 * 90 mm), intended to be inserted between two flat pieces of

the sample to be measured (whose surfaces have been previously rectified) (Fig.3).



Fig. 3. The Ring sonde

The glove box plays a role in conserving changes in the temperature of a sample, the latter consisting of three materials each linked to the other and each giving their function is as follows: a contact thermometer which sets the requested temperature, light bulbs in flames during sample launch in the glove box to heat the sample and then a fan which diffuses the heat in the latter (fig.4).

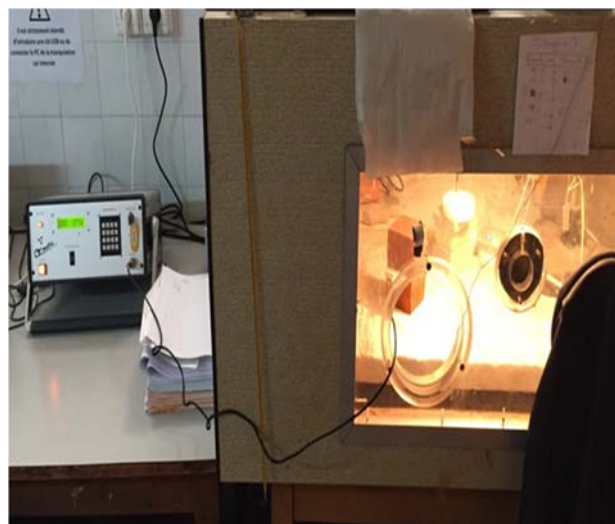


Fig. 4. Glove box

This following table summarizes the mechanical and electrical characteristics of the control unit (table 1).

Characteristics	values
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Power delivered for the ring sonde (w)	From 0 to 2,5
heating time (s)	400
measure time (s)	500
Resistance (ohm)	2,5
Size of the box (mm)	400 x 145 x 260
Weight of the box (Kg)	8
Power supply	230Vac/ 50-60 Hz

Table 1: Mechanical and electrical characteristics of the appliance <<CT-METRE>>

3 Variation of thermal conductivity according to the copper composition

Theory: The equivalent conductivity is calculated according to the composition (x_{cu} , x_{ar}) uses the values executed in Table 2 is considering the material as a binary mixture and the first colon are experimental measures:

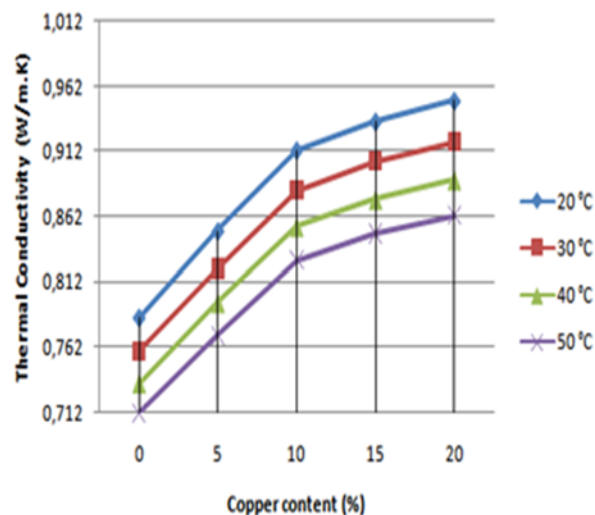


Fig. 5. Influence of thermal conductivity on copper content

Experience: Five pairs of previously prepared samples are taken with different copper content (0%, 5%, 10%, 15%, and 20%) and the temperature in the glove box is set by means of a contact thermocouple successively at (293K, 303K, 313K, and 323K) then we have the measurement of the

thermal conductivity of each pair of sample by introducing it into the CT-metre.

The following values are obtained for the thermal conductivities for each copper content for a given temperature (Table 2):

$x_{Cu}(\%)$	λ à (T=293 K)	λ à (T=303 K)	λ à (T=313 K)	λ à (T=323 K)
0%	0,785	0,741	0,715	0,701
5%	0,851	0,809	0,785	0,762
10%	0,913	0,869	0,831	0,803
15%	0,935	0,909	0,87	0,841
20%	0,951	0,929	0,896	0,862

Table 2: Experimental values of thermal conductivity versus copper content at different temperatures

From the measurements carried out one draws the curves of variation of the thermal conductivity as a function of:

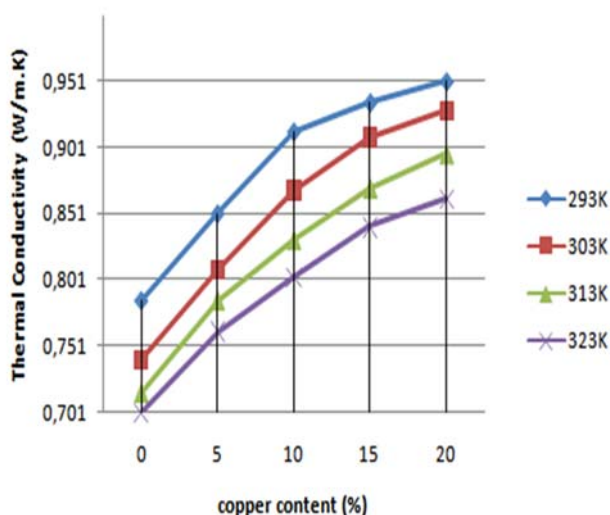


Fig. 6. Experimental influence of thermal conductivity on copper content

Interpretation of the curves

After to the two figs 5 and 6 above theoretical and experimental it is found that the thermal conductivity increases according to the copper content so a perfect agreement between the

theoretical model and the experimental measurements.

The increase of the thermal conductivity as a function of the copper mass content is due to the dipole-charge interactions between the copper atoms and the anionic sites of the clay, these interactions cause the adsorption of copper atoms on the clay and their diffusion through the defects of its crystalline structure. Another factor contributes to the increase of λ which is the heat transfer due to the movement of copper electrons.

4 Variation in thermal conductivity as a function of temperature

Theory: we deduce the relation which reflects the variation of the thermal conductivity as a function of the temperature from the expression of the thermal flux [12].

$$d\Phi = \vec{j} \cdot d\vec{S} = -\lambda \vec{\nabla} T \cdot d\vec{S}$$

Involved $\frac{\Delta\Phi}{\Delta T} = \lambda \frac{S}{L}$

With $\lambda = \Delta\Phi \frac{L}{S\Delta T}$

After the relation above, thermal conductivity decreases with temperature

If $T \gg T_0$

We have $\lambda = \Phi \frac{L}{ST}$

For two different temperatures one can write:

$$\lambda_{ar} = \Phi \frac{L}{ST_{ar}} \text{ And } \lambda_{cu} = \Phi \frac{L}{ST_{cu}}$$

And then we find:

$$\frac{\lambda_{ar}}{\lambda_{cu}} = \frac{T_{cu}}{T_{ar}} \Rightarrow \lambda_{ar} = \lambda_{cu} \frac{T_{cu}}{T_{ar}}$$

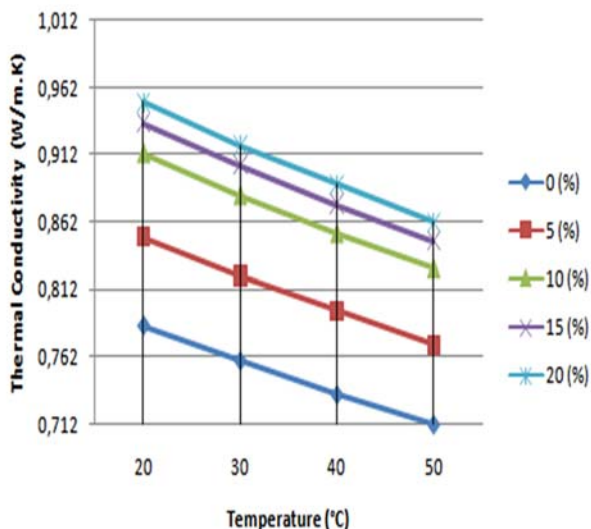


Fig. 7. Influence of the Thermal Conductivity of the Material on Temperature

Experience:

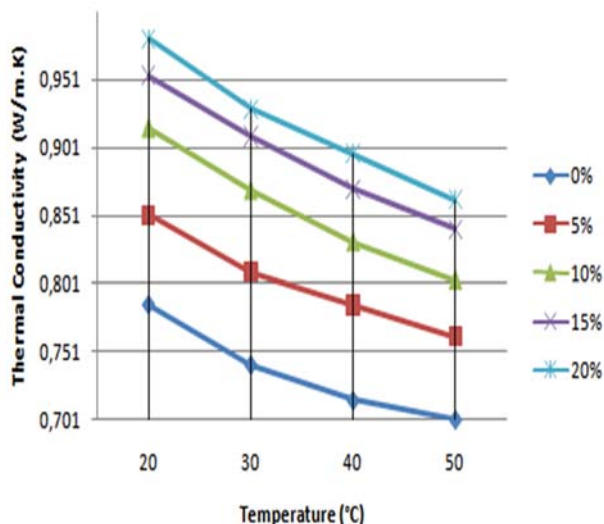


Fig. 8. Experimental influence of the thermal conductivity of the material on Temperature

Interpretation the results obtained:

After the results obtained by the two figs 7 and 8, it can be deduced that the thermal conductivity of the material decreases with the increase of the temperature so λ is a decreasing function with temperature [13].

Since the average free path of the heat-transmitting phonons decreases with temperature due to non-perfect contacts between the two materials as well as the deviations of the crystalline structure of the perfect crystal model clay which cause the phonon attenuation in the material.

5 Conclusion

It has been found from the theoretical model and experimental measurements made by apparatus called CT-METRE, that when doping clay with powdered copper, we end up with a composite material (solid solution of substitution), whose thermal conductivity increases with the copper content and decreases with temperature. This is due to:

- The variation of the conductivity with the copper content, to the increased transfer of heat by electrons in copper as well as the diffusion of copper in the clay and the adsorption of its electropositive atoms on the anionic sites of the clay, which gives it a dual mode of conduction of heat by network and by electrons.
- The variation of the thermal conductivity as a function of the temperature it is due to the interactions of the atoms of the copper with the anionic sites of clay that lead to non-perfect contacts of the two materials therefore to the decrease of the average free path thermal waves, as well as the structural defects existing in the crystalline structure of the clay.

Notation

λ	Thermal conductivity (W/m.k)
T	Temperature (K)
J	Vector heat flow
Φ	Thermal flow
dS	Surface element
x_{cu}	Copper content (%)
x_{ar}	Clay content (%)
L	Length of material (cm)
S	The section (m ²)
λ_{cu}	Thermal conductivity of copper (W/m.K)
λ_{ar}	Thermal conductivity of clay (W/m.K)

Acknowledgment

As authors we would like to thank you very much the Associate Editor for his support and assistance with the review of the paper and all those who participated in this work.

References

[1] S. Pincemin, X. Py, R. Olivès, M. Christ. Highly conductive composites made of

- phase change materials and graphite for thermal storage, *Solar Energy Materials and Solar Cells*, Volume 92, Issue 6, June 2008, Pages 603-613.
- [2] Alberto Torres Miranda , Leandro Bolzoni , Nilam Barekar , Yan Huang , Jesik Shin , SeHyun Ko, Brian John McKay. Processing, Structure and Thermal Conductivity Correlation in Carbon Fibre Reinforced Aluminum Metal Matrix Composites, *Materials & Design*, Volume 156, 15 October 2018, Pages 329-339.
- [3] P. Meukam, A. Noumowe, Y. Jannot et R. Duval, Caractérisation Thermophysique et Mécanique de Briques de Terre Stabilisées en Vue de l'Isolation Thermique de Bâtiment, *Materials and Structures*, Vol. 36, N°261, pp. 453 – 460, 2003.
- [4] P.G. Klemens and D.F. Pedraza, Thermal conductivity of graphite in the basal plane, *Carbon*, Vol. 32, Issue 4, pp 735-741, 1994.
- [5] Francisco Armijo, Francisco Maraver, Manuel Pozo, María Isabel Carretero, Onica Armijo, Miguel Ángel Fernández-Torán, María Virginia Fernández-González, Iluminada Corvillo. Thermal behaviour of clays and clay-water mixtures for pelotherapy, *Applied Clay Science*, Volume 126, June 2016, Pages 50-56.
- [6] Lamine Zerbo, Brahim sorgho, Sié Kam, Julien Soro, Younoussa Millogo, Boubié Guel, Karfa Traoré, Moussa Gomina, Philippe Blanchart, Comportement thermique de céramiques à base d'argiles naturelles du Burkina Faso, *Journal de la Société Ouest-Africaine de Chimie*, 17ème Année, Décembre 2012, N° 034, pages 48- 56.
- [7] R.K. Goyal, K.R. Kambale, S.S. Nene, B.S. Selukar, S. Arbuj, U.P. Mulik. Fabrication, thermal and electrical properties of polyphenylene sulphide/copper composites, *Materials Chemistry and Physics*, Volume 128, Issues 1–2, 15 July 2011, Pages 114-120.
- [8] Y.S. Touloukian, R.W. Powell, C.Y. Ho and P.G.Klemens, Thermophysical properties of matter, *Thermal conductivity of nonmetallic solids*, Vol. 2: IFI/Plenum Press, New-York, 1970.
- [9] A Boulanouar, A Rahmouni, M Boukalouch, A Samaouali, Y Géraud, Mimoun Harnafi, Jamal Sebbani, Determination of Thermal Conductivity and Porosity of Building Stone from Ultrasonic Velocity Measurements, *Geomaterials*, 2013, 3, 138-144
- [10] S. Pincemin, X. Py, R. Olivès, M. Christ, O. Oettinger, Elaboration et caractérisation de matériaux composites de stockage thermique à forte puissance, *Congrès Français de thermique : Défis Thermiques dans l'industrie nucléaire*, 16-19 mai 2006, Ile de Ré.
- [11] Samaouali A, El Rhaffari Y, Hraïta M, Laanab L, Oudrhiri H, Géraud Y, Porous Network Structure and Total Porosity of Rocks Used in Historical Monument Challah (Rabat), *Romanian Journal of Materials*, 2017, 47 (2), 222 -229
- [12] Y. EL Rhaffari , A.Samaouali, L. Laânab, Y. Geraud, M. Boukalouch, Conductivity and thermal diffusivity of the Chellah monument stones, *3rd International Meeting on the Architectural Heritage of the Mediterranean (RIPAM_3)* ,15 - 17 October, 2009 Lusíada University Lisbon, Portugal.
- [13] J.P. Laurent, 'La Conductivité Thermique 'à Sec' des Bétons Cellulaires Autoclavés: un Modèle Conceptuel', *Materials and Structures*, Vol. 24, N°141, pp. 221 – 226, 1991.