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Evaluation of the Electrical Conductivity of Geopolymers Loaded with Carbon Black

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Abstract: Geopolymers are inorganic polymers, composed of aluminosilicates activated with an alkaline solution. This method gives the material three-dimensional networks that are capable of carrying out ion exchange, as well as facilitating the transfer of electric charge through the matrix, when a conductive charge is added. In this sense, geopolymers loaded with carbon black in concentrations of 5, 10, 15 and 20% by weight were produced and the electrical conductivity capacity was verified. The geopolymers were subjected to X-ray Diffraction and resistivity analyzes. The results showed that the geopolymers loaded with carbon black showed a decrease in resistivity due to the concentration of carbon black in the geopolymeric matrix, indicating that with increasing load, less resistive and more conductive. In addition, all analyzes were performed in triplicates, and the calculations of the mean, standard deviation and confidence limit, indicated that in all syntheses the geopolymers remained with the same efficiency.

Keywords: geopolymers, resistivity, conductivity, carbon black, composites.

BJEDIS Scope: This article presents statistical results by ANOVA and Tukey's method, thus verifying the veracity of the results obtained, as well as the significant differences in the analytical data. This is a fundamental step in chemical analysis.

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1. INTRODUCTION

Electrical conductivity is defined as an intrinsic property of materials presenting the ease that free electron flow has to travel within the molecular structure of the material, also defined as conductance specifies conductivity is a quantity that measures the material's ability to carry electrical current in a material. general form is considered as the ratio between the density and the strength of the electric field.

Electrical resistivity is also a property of materials however it is inversely proportional to conductivity the more resistive the material is, the lower its conductivity in relation to the flow of electrical current. The materials can be classified as semiconductor and conductive insulators with order of scale ranging from 10^{-12} ($\Omega.cm^{-1}$), 10^{-6} a 10^{4} ($\Omega.cm^{-1}$) e 10^{5} ($\Omega.cm^{-1}$) respectively. In addition, conductivity is highly influenced by high temperatures, metals that are considered excellent conductors at high temperatures increase their resistivity and consequently decrease their conductivity, in materials considered insulating, the opposite occurs, their resistivity decreases and the material has a conductive behavior (1, 2).

The verification of electrical conductivity is of fundamental importance through it we can check and the best conductive materials to be applied in the daily use of electrical energy. Although metals are excellent conductors, many are composed of metal alloys of different elements, impairing conductivity unlike pure metals such as silver (Ag) and gold (Au) and copper (Cu), which is why it is more viable and commonly applied to pure copper as an electrical conductor. Conductive polymers, also known as intrinsically conductive or semiconductor polymers, are organic composites that have electrical and magnetic properties synthesized from a conductive charge added to an insulating polymer matrix and have as their main advantage their processability through dispersion (2). Geopolymers are inorganic polymers of mineral origin, composed of aluminosilicates activated with an alkaline solution. This method gives the material three-dimensional networks and properties similar to those of Portland cement. Many research seek to incorporate these characteristics of geopolymers to cement in a way that improves its quality by producing new lines of alternative cement, more ecologically efficient through routes that involve less cost. energy and pollution to the environment (3, 4).

The polymeric matrix used in this work is formed by an inorganic geopolymer with properties similar to cement, among them its high resistivity, being characterized as an insulating material for its electrical applicability, it was necessary to modify the geopolymer structure by adding conductive magnetic charges. copper and aluminum for being excellent conductors, however due to the high density of copper and the high oxidation of aluminum, it hindered the synthesis of the compound. That is why I was used in this work nano carbon black particles acting as a conductive additive because it is a material of diverse industrial application mainly in the paint and rubber industry and is abundant in carbon which allows its four electrons in the valence layer to pose link their atoms together to form compounds of totally different structures and properties (5).

The electrical conductivity of carbon black is high according to its structure, surface area and composition before functional groups (5, 6). Thus, its electrical conductivity will be greater according to the increase in the number of concentration and interaction between its particles in the polymeric matrix, allowing a path that performs the transport of the electric current in the medium should be as low as possible in a way that preserves the characteristics and properties of the polymeric matrix and can adjust it for the intended application.

The biggest challenge in this work is to find a route that allows the synthesis of the material by dispersing and distributing the charge, in the polymeric material allowing the conduction of the electric current and reducing the charge concentration. This work aims to evaluate the carbon black ability to decrease the resistivity of composites produced from geopolymeric matrices. In addition, verify the crystalline composition using the X-Ray Diffraction technique.

2. METHODOLOGY

2.1. Production of conductive composites

To produce the composites, Printex conductive carbon black, sodium silicate P.A, sodium hydroxide P.A and metakaolin were used. Production followed in two stages:

The first step consisted of weighing with the aid of a 150 mL beaker and an analytical balance, 2.6 g of sodium hydroxide, 6.15 g of sodium silicate. Then, 13 ml of deionized water were collected and added in a 100 ml beaker, the sodium silicate and sodium hydroxide were then inserted into the water and stirred at 300 rpm, until total solubilization. After 4.5 g of metakaolin was added to the alkaline solution and carbon black (CB), in concentrations of 5%, 10%, 15% and 20% by weight, the agitation lasted for 5 minutes until the total homogenization of the carbon black to the geopolymer.

The second step consisted of molding the material, using a 7.5 cm polyethylene tube, after which the material was placed to cure at room temperature (35 °C) for 2 days. All syntheses were performed in triplicate to verify the similar behavior in all samples in the analyzes that were subsequently submitted.

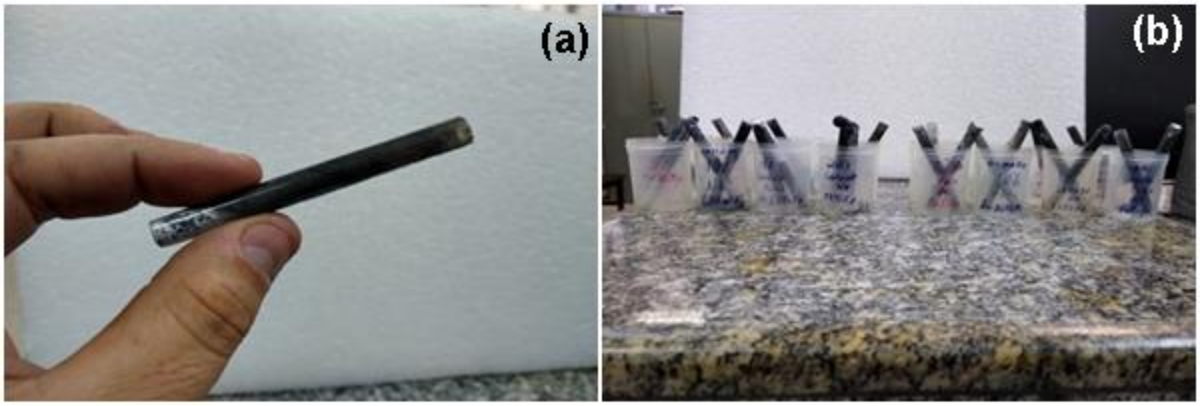


Figure 1. Illustration of the mold in a polyethylene tube (a) and samples molded in triplicates (b)

2.2. Characterizations

2.2.1. X-Ray Diffraction

X-ray diffraction tests (XRD) were performed on a Rigaku X-ray diffractometer, model Miniflex. The method used was FT, in an angular step of 0.05º, in the range of 2θ between 10 ° to 80 °, with acquisition time in each point of 1.0s**.**

2.2.2. Resistivity test (Ω)

Measurements were obtained with a homemade sample holder coupled to a Keithley 6517-B electrometer. The average diameter of the sample holder was 0.1350.03 mm. The total sampling time and electrification time were equal to 120 s. The tests were performed in triplicates.

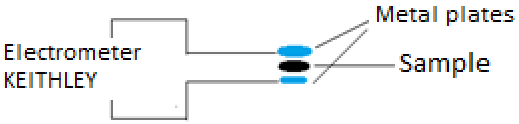


Figure 2. Illustration of the resistivity measurement method.

2.2.3. Resistivity

Resistivity was measured using the formula already described in the literature (11).

$$\rho = \frac{(R * \pi * r)^2}{e} \qquad (1)$$

Where:

ρ = resistivity ($\Omega.cm^{-1}$)

R= resistance (Ω)

r= radius (cm)

e= sample thickness (cm)

2.2.4. Statistical Analysis

The average values of the variables obtained in the treatments were compared using Tukey's test at 5% probability using the SigmaPlot 12.5 program (7, 8).

3. RESULTS AND DISCUSSION

The geopolymers were characterized by the X-Ray Diffraction technique in order to investigate the crystalline property of the material and the presence of the magnetic particle. The results obtained showed that, the peaks of the main components of the geopolymer formation were identified, being these, sodalite (Na_8 ($AlSiO_4$) 6 (NO_2) $_2$) at angles 37 ° and quartz (SiO_2) at angle 40 °. It was also possible to identify the characteristic peak of carbon black at 43 ° (9–11). These results show that the formation of the geopolymer and the production of the composites were carried out. In addition, it was possible to investigate that, as the carbon black is increased in the matrix, the crystallinity of the composites increases.

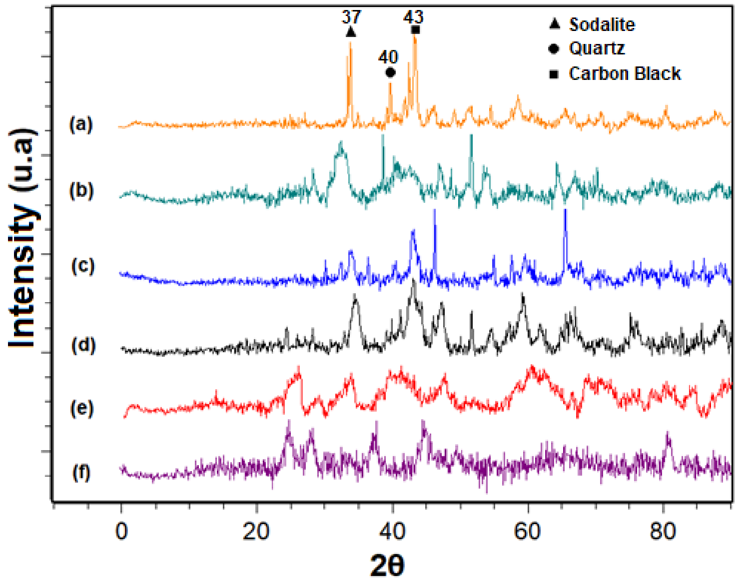
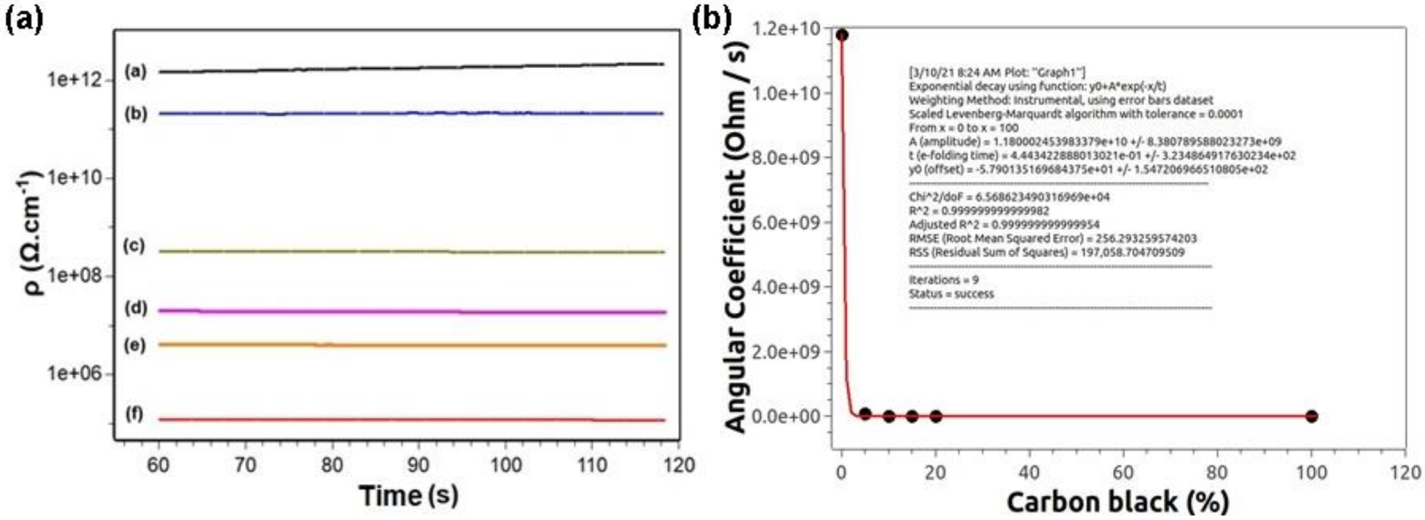


Figure 3. Diffractogram of composites (a) 20% carbon black, (b) 15% carbon black, (c) 10% carbon black, (d) 5% carbon black, (e) geopolymer and (d) carbon black.

The geopolymer presents a linear decrease of the electric resistance versus time due to its polarizable nature under a constant electric field.

Carbon black (CB) presents the opposite behavior, already reported in the literature (12). This behavior is due to overcoming the carbon black resistive surface barrier, composed of oxygen-rich-species and adsorbed oxygen. With the increase of the electrification time, the CB surface's polarization drives the electric current into the particles, composed almost exclusively of carbon, increasing the observed electric conduction. Besides that, there is evidence of a percolative phenomenon. After 5% of the conducting filler, the CB increase inside the matrix produces an electrical behavior similar to that presented by the pure carbon black (Figure 4 (a)). Also, the drop of the resistance over time assumes a linearly progressive aspect, indicating that the matrix's alkaline nature favors the CB surface's polarization, making the intraparticle charge transportation easier. Finally, the slope values drop can be modeled by exponential decay, as shown in Figure 4 (b). The value of the calculated e-folding can be considered as tending to zero. Thus, the experimental evidence shows that the matrix can modify the physical nature of the filler. Besides, small amounts of CB inside the geopolymeric matrix produce drastic changes in the composite material's electrical nature.



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[3/10/21 8:24 AM Plot: "Graph1"]
Exponential decay using function: y0+A*exp(-x/t)
Weighting Method: Instrumental, using error bars dataset
Scaled Levenberg-Marquardt algorithm with tolerance = 0.0001
From x = 0 to x = 100
A (amplitude) = 1.180002453983379e+10 +/- 8.380789588023273e+09
t (e-folding time) = 4.443422888013021e-01 +/- 3.234864917630234e+02
y0 (offset) = -5.790135169684375e+01 +/- 1.547206966510805e+02
------------------------------------------------------------
Chi^2/doF = 6.568623490316969e+04
R^2 = 0.999999999999982
Adjusted R^2 = 0.999999999999954
RMSE (Root Mean Squared Error) = 256.293259574203
RSS (Residual Sum of Squares) = 197,058.704709509
------------------------------------------------------------
Iterations = 9
Status = success
------------------------------------------------------------
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Figure 4. Figure 4 (a): Resistivity of (a) geopolymer, (b) composite with 5% CB, (c) composite with 10% CB, (d) composite with 15% CB, (e) composite with 20% CB and (f) CB. Figure 4 (b): Angular coefficient of geopolymer and composites.

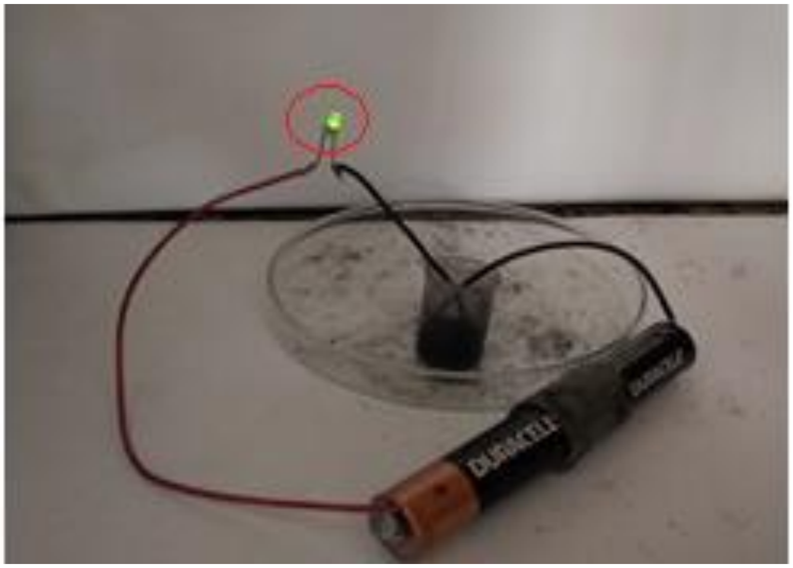

Figure 6. Illustration of conductivity test of the material using an electric current.

Table 1. Average values as a function of different treatments of the geopolymer, carbon black and composites with 5%, 10% ,15% and 20%. Means followed by the same lowercase letter do not differ statistically from each other by the Tukey test at 5% probability.

Materials	**ρ (Ω.cm^{-1})** **Average**
Geopolymer	$2{,}14\times10^{12} \pm 1{,}79\times10^{6\ a}$
CB	$1{,}19\times10^{5} \pm 6{,}62\times10^{2\ b}$
5%	$2{,}10\times10^{11} \pm 2{,}63\times10^{5\ c}$
10%	$3{,}12\times10^{8} \pm 6{,}66\times10^{3\ d}$
15%	$1{,}88\times10^{7} \pm 8{,}85\times10^{2\ e}$
20%	$3{,}99\times10^{6} \pm 2{,}72\times10^{2\ f}$

Means followed by the same lowercase letter in the column do not differ statistically from each other by the Tukey's test at 5% probability.

4. CONCLUSION

Through the characterization of the DRX and the resistivity tests applied in these composites, the conductive capacity of the geopolymer loaded with carbon black particles was proven, being characterized as a semiconductor material of simple preparation and low cost that increases its electrical conductivity according to the quantity and charge inserted in the polymeric matrix due to the presence of free ions in the structure. In addition, through a simple test using an electric current system with a battery, the material proved to be efficient when lighting a led lamp, and the statistical results showed that the carbon black had a significant difference in geopolymeric matrix, decreasing the resistivity of composites. This result is very encouraging for future studies and application for new alternative drivers.

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Sample CRediT author statement

Fabíola da Silveira Maranhão: Conceptualization, Methodology, Data analysis, and Writing-Original draft preparation. **Fernando G. de Souza Junior**: Conceptualization, Supervision, Reviewing and Editing. **Tiago Galdino Silva da Costa**: Data analyses and Reviewing, **Cintia Patrícia Santos da Paixão**: Reviewing, **Sérgio Thode Filho**: Conceptualization, Supervision, Reviewing and Editing, **Diganta B. Das**: Reviewing, Editing and Validation

REFERENCES

1. Condutores e isolantes: o que são, diferenças, exemplos. **Brasil Escola [online]**. Accessed 31 March 2021. Available from: https://brasilescola.uol.com.br/fisica/condutores-isolantes.htm.

2. ARULRAJAH, Arul, KUA, Teck-Ang, HORPIBULSUK, Suksun, PHETCHUAY, Chayakrit, SUKSIRIPATTANAPONG, Cherdsak and DU, Yan-Jun. Strength and microstructure evaluation of recycled glass-fly ash geopolymer as low-carbon masonry units**. Construction and Building Materials**. v. 114, p. 400–406. 2016. DOI 10.1016/j.conbuildmat.2016.03.123.

3. ASSI, Lateef N., DEAVER, Edward (Eddie), ELBATANOUNY, Mohamed K. and ZIEHL, Paul. Investigation of early compressive strength of fly ash-based geopolymer concrete**. Construction and Building Materials**. v. 112, p. 807–815. 2016. DOI 10.1016/j.conbuildmat.2016.03.008.

4. BIONDI, Lorena, PERRY, Marcus, MCALORUM, Jack, VLACHAKIS, Christos and HAMILTON, Andrea. Geopolymer-based moisture sensors for reinforced concrete health monitoring**. Sensors and Actuators B: Chemical**. v. 309, p. 127775. 2020. DOI 10.1016/j.snb.2020.127775.

5. WANG, Meng-Jiao, GRAY, Charles A., REZNEK, Steve A., MAHMUD, Khaled and KUTSOVSKY, Yakov. Carbon Black. In : **Kirk-Othmer Encyclopedia of Chemical Technology** [online]. American Cancer Society, 2003. Accessed 31 March 2021. ISBN 978-0-471-23896-6. Available from: https://onlinelibrary.wiley.com/doi/abs/10.1002/0471238961.0301180204011414.a01.pub2.

6. PANTEA, Dana, DARMSTADT, Hans, KALIAGUINE, Serge and ROY, Christian. Electrical conductivity of conductive carbon blacks: influence of surface chemistry and topology**. Applied Surface Science**. v. 217, n. 1, p. 181–193. 2003. DOI 10.1016/S0169-4332(03)00550-6.

7. COSTA, NETO and ESTÁTISTICA, P. L. O. 2a Edição**. São Paulo: Editora Edgard Blücher**. 2002.

8. MILLER, J. C., MILLER, J. N. and WORSFOLD, P. J. Statistics for analytical chemists**. Analytica Chimica Acta**. v. 184, p. 337–338. 1986.

9. ZHENG, Zhao, MA, Xue, ZHANG, Zuhua and LI, Yuxiang. In-situ transition of amorphous gels to Na-P1 zeolite in geopolymer: Mechanical and adsorption properties**. Construction and Building Materials**. v. 202, p. 851–860. 2019. DOI 10.1016/j.conbuildmat.2019.01.067.

10. AWADH, Salih Muhammad and YASEEN, Zaher Mundher. Investigation of silica polymorphs stratified in siliceous geode using FTIR and XRD methods**. Materials Chemistry and Physics**. v. 228, p. 45–50. 2019. DOI 10.1016/j.matchemphys.2019.02.048.

11. R. OSTYN, Niels, A. STEELE, Julian, PRINS, Michiel De, PULINTHANATHU SREE, Sreeprasanth, VINOD CHANDRAN, C., WANGERMEZ, Wauter, VANBUTSELE, Gina, WON SEO, Jin, J. ROEFFAERS, Maarten B., BREYNAERT, Eric and A. MARTENS, Johan. Low-temperature activation of carbon black by selective photocatalytic oxidation**. Nanoscale Advances**. v. 1, n. 8, p. 2873–2880. 2019. DOI 10.1039/C9NA00188C.

12. DING, Tianhuai, WANG, Luheng and WANG, Peng. Changes in electrical resistance of carbon-black-filled silicone rubber composite during compression**. Journal of Polymer Science Part B: Polymer Physics**. v. 45, n. 19, p. 2700–2706. 2007. DOI https://doi.org/10.1002/polb.21272.