Synthesis, Thermal Properties and Electrical Conductivity of Na-Sialate Geopolymer

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Received: 25 January 2022 / Accepted: 1 April 2022 / Published online: 21 April 2022
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Abstract
This work aims to study the thermal behavior of basic-geopolymers derived from metakaolin (clay). The geopolymers were characterized by different techniques: thermal analysis (DTA, TGA), X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) and impedance spectroscopy. Some physicochemical properties of the products were also determined: the phases obtained after geopolymer heat treatment and their electrical properties. The results obtained after drying and heat treatment showed that the products kept their initial shapes, but revealed variable colors depending on the temperatures at which they were treated. The products obtained are amorphous between 300 up to 600 °C with peaks relating to the presence of nanocrystallites of muscovites and zeolite, thus at 900 °C it is quite amorphous but only contains nanocrystallites of muscovites. From the temperature of 950 °C, we notice that the geopolymer has been transformed into a crystalline compound predominated by the Nepheline (NaAlSiO4) with the presence of a crystalline phase by minor peaks of Muscovite, this crystalline character has been increased at 1100 °C to obtain a whole phase crystalline of a Nepheline. The treatment of this geopolymer for one hour at 1200 °C shows an amorphous phase again corresponding to corundum (α-Al2O3). This indicates that the dissolution of the grains by the liquid phase induces the conversion of the material structure from sialate [–Si–O–Al–O] to sialate siloxo [–Si–O–Al–O–Si–O–] and the formation of a new crystalline phase (α-Al2O3). This development of sialate to sialate-siloxo was confirmed by IR spectroscopy. As mentioned above, from 300 to 900 °C, Na-sialate geopolymer exhibits the same disorder structure of nepheline. The crystal structure of nepheline is characterized by layers of six-membered tetrahedral rings of exclusively oval conformation. The rings are built by Regularly alternating tetrahedral AlO4 and SiO4. Stacking the layer’s parallel to the c axis gives a three-dimensional network containing channels occupied by Na cations. This topology favors easy movement of Na+ ions throughout the structure. For this reason, ionic migration in nepheline is widely reported. The refinement of Na-Sialate geopolymer at room temperature gives bulk high ionic conductivity of about 5 × 10−5 S cm−1 and this is due to the probable joint contribution of H+ and Na+ ions. Above 200 °C, Na+ seems to remain the only charge carrier with a low activation energy of about Ea = 0.26 eV. At higher temperatures, the characteristic frequencies become so close that it is impossible to distinguish the contributions. A total resistance comprising both grain and grain boundaries contribution is then determined.

Keywords Na-Sialate geopolymer · Nepheline · Thermal analysis · Electrical Conductivity · IR spectroscopy

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1 Introduction

Nanomaterials have generated extraordinary research interest due to their unique and beneficial applications in different fields. One of the nanoscale oxides is geopolymer which is considered an important oxide due to its good application performance and various modification mechanisms [1–11].

As well as their potential practical applications based on its high temperature refractory properties. During preliminary experiments with polysialate geopolymers, we observed a high degree of thermal stability of these materials, particularly potassium polysialates, some of which appeared to have melting points as high as 1400 °C. Some of its potential uses include the manufacture of solid-state batteries and electrochemical sensors that has causes of its high level of electrical conductivity which should play an important role in fast ionic conduction thanks to free sintering and low cost [12].

Inorganic geopolymers are materials with ceramic properties but which are synthesized and hardened by curing at room temperature. Due to the low energy requirements of their production from raw materials (clays) and their high flammability at high temperatures, these compounds are attracting increasing interest as environmentally friendly fireproof building materials [13, 14]. Sound insulation and materials to encapsulate hazardous substances, waste for storage or disposal.

So far, different routes have been presented to prepare a geopolymer of great importance for potential studies of its properties. It has been shown that the size and shape of the particles of nanomaterials have a significant impact on their characteristics. Thus, it is important and necessary to explore appropriate methods to prepare a base geopolymer and control its morphology and particle size.

At present, we are witnessing intensive research in the field of geopolymer synthesis and the preparation of new technically applicable materials from these geopolymers [15–18]. As is known, the basic raw materials used for the synthesis of geopolymers are aluminosilicate minerals such as kaolinite, albite, stilbite [19, 20]. These raw materials must be transformed into a reactive form by several hours of heating at approximately 750 °C before the synthesis of the geopolymers. This process, called basic raw material activation, requires significant energy consumption, or these materials are usually made from clays activated by an alkali or an alkali-silicate solution [13, 14, 21]. When processed below 100 °C, they are generally X-ray amorphous and consist of cross-linked tetrahedral AlO4 and SiO4. The arrangement of the silicon and aluminum tetrahedron in a three-dimensional network structure generates cavities in which the water molecules and the monovalent cations (Na+, and/or K+ type) will be trapped.

The proposed empirical formula is: $\text{M}_n((–\text{SiO}_2)_z–\text{AlO}_2)_n·w\text{H}_2\text{O}$ where M is a positive cation, such as sodium or potassium, $z$ is 1, 2 or 3, $n$ is the degree of polycondensation and $w$ is the number of moles of hydrated water (usually up to 7) [13].

In the present work, the structural characterizations, the thermal evolution and the ionic conductivity of the new Na-Sialate geopolymer were studied by different experimental studies.

2 Materials and Experimental Techniques

2.1 Geopolymer Preparation

The Na geopolymer was synthesized from well-cry stallized commercial kaolin. The thermal activation of the kaolin was carried out at 700 °C for 6 h (to obtain a metakaolin). The chemical composition of kaolin was determined by X-ray fluorescence with Thermo Nitron FXL. A sodium hydroxide solution used in this study was a commercial base NaOH 85%, whose molar ratios Si/Al = 1 and Na/Al = 1. The chosen ratio of H2O/NaOH of the sodium hydroxide solution was 10.

The geopolymer was prepared by mechanically mixing distilled water, stock solution and metakaolin to form a homogeneous suspension. The present work was performed using a previously found composition to prepare the Na-PS formulation [22, 23].

From the first minutes, a paste was obtained and evolved chemically and rheologically very quickly. This paste was composed of a gel solution and an interstitial liquid. After several hours it would harden to form an amorphous solid.

The sample was kept at room temperature for 7 days and then calcined at different temperatures.

2.2 Experimental Techniques

2.2.1 X-Ray Powder Diffraction

The phase identification of the geopolymer was performed by X-ray diffractometry (BRUKER-AXS-D8-Advance) with CoKα radiation ($\lambda = 0.178$ nm) on geopolymer heated at different temperatures and a scanning rate of 10°/min from 10° to 60° (2θ) (step = 0.066, time/step = 100 s), the same type of diffractometer working with CuKα radiation ($\lambda = 0.154$ nm) on a PANalytical X’pert Pro diffractometer equipped with the X’celerator detector in the 2θ range from 5 to 60 (step = 0.033, time/step = 50 s) was used to analyze the kaolin, metakaolin and unheated geopolymer.
2.2.2 IR Spectroscopy

The IR measurements, obtained using a Perkin Elmer 1600 FT spectrometer, were carried out at ambient temperature between 400 and 4000 cm\(^{-1}\).

2.2.3 Thermal Analysis

The thermal behaviors were characterized by differential thermal analysis and thermogravimetry (TG–DTA). This analysis was performed by Instrument: SDT Q600 V8.1 Build 99, the temperature was raised from room temperature up to 1400 °C at a heating rate of 10 °C/min under dry air flow (100 mL·min\(^{-1}\)) with \(\alpha\)-Al\(_2\)O\(_3\) as reference.

2.2.4 Electrical Conductivity

Electrical conductivity measurements were performed on geopolymer pellet sintered at 300 °C. Specimen pellet was produced with a diameter \(\approx 5\) mm and thickness \(\approx 2.4\) mm. Thin platinum films (~ 10 nm) were deposited on each face of the pellet by radiofrequency sputtering using a Sputter Coater Polaron SC7620 apparatus. The impedance spectra were recorded over the frequency range from 1 MHz to 1 Hz every 50 °C from 75 to 725 °C, under dry air flow, using a Schlumberger Solartron 1260 frequency response analyzer connected to a Schlumberger Solartron 1296 dielectric interface. An ac voltage of 350 mV was used, and a waiting time of 20 min was necessary to reach thermal equilibrium after each 50 °C heating or cooling step.

Equivalent circuits modeling was used to analyze the impedance spectra and the corresponding parameters were obtained by complex nonlinear least squares (CNLLSQ) fitting [24] performed on commercial software (Zview) [24].

3 Results and Discussion

3.1 Characterization of Raw Material

3.1.1 X-Ray Powder Diffraction

The chemical composition of the kaolin, determined by X-ray fluorescence, is reported in Table 1, the clay was rich in silica, alumina and contained an amount of iron, calcium, magnesium and potassium oxides indicating the probable presence of muscovite or illite, in the 2:1 phyllosilicate, Al\(^{3+}\) in the octahedral sheets was replaced with Mg\(^{2+}\) and Fe\(^{3+}\), and K\(^{+}\) in the interlayer spaces was replaced with Na\(^{+}\) and Ca\(^{2+}\).

The XRD pattern of kaolin (Fig. 1) was analyzed by Le Bail method, without atoms, using the pattern matching routine of the FULPROF program [25]. The profile refinement results indicate that the diffraction pattern corresponds to a mixture of two phases: Kaolinite (C 1; \(a = 5.055(1)\) Å, \(b = 8.984(5)\) Å, \(c = 7.376(4)\) Å, \(\alpha = 91.730(6)^\circ\), \(\beta = 104.92(3)^\circ\), \(\gamma = 89.67(4)^\circ\), and Muscovite (C 2/c; \(a = 5.180(2)\) Å, \(b = 8.932(3)\) Å, \(c = 20.034(3)\) Å, \(\alpha = 90.0^\circ\), \(\beta = 95.14(8)^\circ\) and \(\gamma = 90.0^\circ\)). The commercial kaolin is a mixture of 94% of kaolinite and 6% of Muscovite.

3.1.2 IR Spectroscopy

3.1.2.1 Crystal Structures  Kaolinite with general formula: Al\(_2\)(OH)\(_4\)Si\(_2\)O\(_5\) is 1:1 type layer silicates and consists of a two-layer arrangement of tetrahedral and octahedral sheets. The tetrahedral layer is composed of SiO\(_4\) linked in a hexagonal array. The bases of the tetrahedral are approximately coplanar and the apical oxygen atoms are linked to a second layer containing aluminum ions and OH groups. The aluminum ions located in the center of octahedral are surrounded by 4(O) and 2(OH) [26].

A major component of the separation force is the hydrogen-bonding force between hydroxyl groups on octahedral sites and oxygen atoms of SiO\(_4\) tetrahedral, as van der Waals force among the atoms.

![Fig. 1](image)

**Table 1** Chemical composition of kaolin (% of mass)

|         | SiO\(_2\) | Al\(_2\)O\(_3\) | FeO\(_3\) | MgO | Na\(_2\)O | CaO | K\(_2\)O |
|---------|-----------|----------------|-----------|-----|-----------|-----|---------|
| 49.17   | 44.05     | 0.69           | 0.27      | 1.43| 0.81      | 2.13|         |

The point symbols represent the observed diffraction pattern, the solid lines represent the calculated pattern, and the curves at the bottom of the figure represent the difference between the observed and calculated patterns. The short vertical lines mark the positions of possible Bragg reflections.
3.1.2.2 Unit Cell OH Stretching Vibrations

The identification of proton positions in the structure of layer silicate are important to interpret infrared spectroscopy data of kaolin.

The kaolinite contains four (OH) groups in the primitive unit cell. The observed band at 3620 cm\(^{-1}\) can be assigned to the (OH) stretching vibration of one hydroxyl group which lies within the layers and is nearly parallel to the layer surface, a (001) plane [27, 28]. The other three (OH) groups, lie on the upper surface of the layers, and are oriented at angles of 60–73° to the (001) plane [27, 28]. The three hydroxyl groups couple to give a strong in-phase symmetric stretch, observed, in our case, at 3691 cm\(^{-1}\) and two weak out-of-phase vibrations at 3669 cm\(^{-1}\) and 3652 cm\(^{-1}\) [29].

The IR spectrum of kaolin (Fig. 2) exhibits additional bands at 3677, 3727, 3730, 3740 cm\(^{-1}\) in the (OH) stretching region that are not reported at the previous research works. These extra bands were observed in the Raman spectrum of kaolinite at 3686 and 3710 cm\(^{-1}\) [28], and assigned to transverse optical modes involving in-phase coupled vibrations of the layer-surface hydroxyl groups. Farmer suggested that the corresponding IR bands have transition moments nearly perpendicular to the layer surface, and appear in the higher frequencies range of the longitudinal optical modes of macroscopic crystals.

3.1.2.3 Vibrations Below 1200 cm\(^{-1}\)

The infrared vibration bands of kaolin below 1200 cm\(^{-1}\) displayed in Fig. 3, represent cooperative motions of structural ionic groups. The IR vibration bands may be divided into vibrational, rotational and translational motions of anion-cation associations. Although, the proportion of each factor in the production of vibrations is difficult to evaluate at the present time. That is why the detailed vibrational assignments of bands in this region are controversial. To reveal the general characteristics of these network vibrations we have adapted assignments that present a general agreement.

The moderately strong broad band at 1033 cm\(^{-1}\) and the strong sharp band at 1004 cm\(^{-1}\) correlated to the asymmetric and symmetric stretching modes of Si–O–Si, respectively [30]. A medium band appeared at 1117 cm\(^{-1}\) correspond to the apical Si–O vibrations [30–32]. The weak bands at 940 and 912 cm\(^{-1}\) assigned to the deformation modes of inner and inner-surface of Al\(_2\)O–H [31, 32].

The domain observed from 800 to 400 cm\(^{-1}\) is the skeleton vibrational modes region. There is a general agreement that the medium band observed at 428 cm\(^{-1}\), the
weak band at 650 cm\(^{-1}\) and the strong bands at 460 and 528 cm\(^{-1}\) belong to Si–O–Si deformation banding modes [31, 32].

Three medium bands appeared at 688, 752 and 795 cm\(^{-1}\) were assigned to the deformation modes of Al\(^{VI}\)-O–Si bands [31, 32].

A weak band near of 413 cm\(^{-1}\) ascribed to out-of-plane OH liberations [33].

### 3.2 Characterization of Na-Geopolymer

#### 3.2.1 X-Ray Diffraction

The Diagrammes of DRX of metakaolin, geopolymer at room temperature and geopolymer heated for 1 h at 300, 600 and 900 are shown in Fig. 4.

Metakaolin comes from the calcination at 700 °C of kaolin. Metakaolin exhibits a relatively amorphous XRD pattern with notable crystalline peaks compared to muscovite phase nanoparticles. This is characteristic of the potentially reactive phase of geopolymerization.

The XRD model of the geopolymer showed the typical amorphous structure of metakaolin. Moreover, the diffraction peaks characteristic of muscovite remained after geopolymerization, suggesting that muscovite did not participate in the geopolymerization reaction. While the amorphous structure of metakaolin transformed from one structure to another structure.

A broad peak characteristic of an amorphous structure with two peaks corresponding to muscovite and zeolite nanoparticles was observed at 300 °C and persisted up to 600 °C.

After treatment at 900 °C, the nanocrystallites of muscovite remained stable, while the zeolite disappeared (Fig. 5; Table 2).

After heating at 950 °C, Na-geopolymer transformed to crystalline material consisting predominantly in nepheline (NaAlSiO\(_4\)) with a few low intensities, minor peaks are ascribable to the muscovite crystalline phase. On heating at 1100 °C, the amount of the amorphous phase increases with the decrease of crystalline peaks intensities of the nepheline phase. The relative decrease should be ascribed to the appearance of the liquid phase in the matrix. The liquid will affect crystalline phases by partial dissolution (Fig. 6).

At 1200 °C, diffraction lines corresponding to corundum (\(\alpha\)-Al\(_2\)O\(_3\)) are superimposed on the broad amorphous background feature (Fig. 6). These indicates that the dissolution of grains by liquid phase induces the conversion of the structure of the material from sialate [–Si–O–Al–O] to sialate siloxo [–Si–O–Al–O–Si–O–] and the formation of new crystalline phase (\(\alpha\)-Al\(_2\)O\(_3\)). This development of sialate to sialate-siloxo was confirmed by IR Spectroscopy.

#### 3.2.2 IR Spectroscopy

Figure 7 illustrates the infrared spectra of kaolin, metakaolin and geopolymer treated at different temperatures for 1 h.

The IR spectrum of kaolin showed a dihydroxylation phenomenon during its transformation to metakaolin correlated by the appearance of peaks near of 3650 cm\(^{-1}\).

On the IR spectrum of metakaolin, a peak appeared at 1037 cm\(^{-1}\) corresponds to a large concentration of Si–O bonds. Two infrared bands are located at 569 cm\(^{-1}\) and 648 cm\(^{-1}\) assigned to Si–O–Al stretching vibrations; a peak at 439 cm\(^{-1}\) arises from Si–O–Si bending vibration. Indeed, an IR band observed at 806 cm\(^{-1}\) is assigned to the Al–O bonding mode of [AlO\(_4\)] octahedra [30, 31].

Due to the presence of water molecules in geopolymer, the strong characteristic peaks at approximately 3331 cm\(^{-1}\) and 1631 cm\(^{-1}\) were attributed to stretching and bending
vibrations of hydroxyl groups, respectively [32, 33]. After geopolymerization, the chemical environment around regular arranged chain structures of the Si–O bond altered, along with the formation of Al–O–Si bonds. Subsequently, the strong asymmetrical stretching vibration peak of the Si–O bond on the IR spectrum of metakaolin (1037 cm$^{-1}$) shifted to a lower wavenumber (960 cm$^{-1}$). This indicated that the solidification process of the geopolymer is a chemical reaction, according to a generation of new substance.

The principal new band appeared at 960 cm$^{-1}$ is assigned to the asymmetric stretching vibration of Si–O–T band links in the geopolymer frameworks (T:Si or Al in tetrahedral coordination). This band is known to be sensitive to connectivity and Si/Al ratio [34, 35], and in this case, it is observed at a wavenumber consistent with the presence of predominantly Si–O–Al bonds, which agrees well with the stoichiometry showed in previously studied systems [34, 35].

It was also found that the broad and strong peak observed at approximately 802 cm$^{-1}$; which belongs to the stretching vibration of hexa-coordinate Al(VI)–O in metakaolin; almost disappeared after geopolymerization.

A new peak located at 697 cm$^{-1}$ corresponds to the bending vibration of tetra-coordinated Al(IV)–O–Si in a cyclic structure emerged on the FT-IR spectra of the

| Table 2 | The wavenumber of kaolin, metakaolin and geopolymer treated at different temperature |
|---------|-----------------------------------------------|
| Kaolin  | Metakaolin | Geopolymer | 100 °C | 300 °C | 900 °C | 1100 °C | 1200 °C |
| 3740    | –          | 3331       | 3344   | 3347   | –      | –       | –       |
| 3730    | –          | 1631       | 1647   | 1654   | –      | –       | –       |
| 3727    | –          | –          | 1564   | 1549   | –      | –       | –       |
| 3677    | –          | –          | 1379   | 1371   | –      | –       | –       |
| –       | –          | –          | –      | –      | 1221   | 1367    | –       |
| –       | –          | –          | –      | –      | –      | –       | –       |
| 1117    | –          | –          | –      | –      | –      | –       | 991     |
| 1033    | 1037       | 960        | 962    | 968    | 975    | 970     | 846     |
| 1004    | –          | 843        | 853    | 848    | –      | –       | –       |
| 940     | –          | –          | –      | –      | –      | –       | –       |
| 912     | 806        | –          | 716    | 729    | 708    | –       | 699     |
| –       | –          | 697        | –      | –      | –      | 682     | 636     |
| 795     | –          | 545        | –      | –      | –      | 654     | 574     |
| 752     | 648        | 470        | 558    | 572    | 555    | 556     | 442     |
| 688     | 569        | 421        | 462    | 467    | 456    | 465     | –       |
| 650     | –          | –          | –      | –      | –      | –       | –       |
| 528     | 439        | –          | –      | –      | –      | –       | –       |
| 460     | –          | –          | –      | –      | –      | –       | –       |
| 413     | –          | –          | –      | –      | –      | –       | –       |

Fig. 6 Typical X-ray powder diffractogram of geopolymer heated for 1 h at 1200 °C (* α-Al$_2$O$_3$)

Fig. 7 FT-IR spectra of kaolin, metakaolin and geopolymer cured at different temperatures
geopolymers. This phenomenon signified the formation of aluminosilicate networks with the transition from hexacoordinated Al(VI) to tetra-coordinated Al(IV) during the geopolymerization process, as observed by Sitarz et al. [36].

After heating at 100 °C, the geopolymer produces a marked decrease in the water bands (Fig. 7).

Apart from this obvious change in the sample hydration on heating to 1100 °C, small changes also occur in the Al–Si–O region, especially the development of the broad zeolites band at about 725 cm⁻¹ (see Fig. 7, T = 300 and 600 °C, consistent with the XRD results).

In summary, all the results indicate that the removal of hydration water from a well-treated Na-PSS1 polymer brings about no significant change to the structure. Furthermore, the structure displays a high degree of thermal stability at higher temperatures, retaining its X-ray amorphous character and the atomic environment of its constituents, Al, Si anéd Na components.

Finally, on the spectrum of 1200 °C a double band observed at 635 and 592 cm⁻¹ (Fig. 7) relative to the presence of crystalline corundum (α-Al₂O₃) phase [37] (already detected through XRD). It supports a hypothesis of the conversion of the structure of the material from sialate [–Si–O–Al–O] to sialate-siloxo [–Si–O–Al–O–Si–O–] according to the appearance of α-Al₂O₃ crystalline phase. The principal band at 960 cm⁻¹ characteristics of sialate shifted to a lower wavenumber (846 cm⁻¹). The principal new band at 846 cm⁻¹ is consistent with the presence of sialate-siloxo [34, 35].

3.2.3 Thermal Analysis by TG–DTA

The thermal analysis curves TG and DTA of Na-geopolymer are shown in Fig. 8.

Below 250 °C, the TG curve indicates that the sample present about 14% of mass loss related to the evaporation of free water (~70%). This phenomenon is well evidenced on the DTA thermogram by the presence endothermic peaks appearing from ambient temperature until approximately 250 °C with a minimum values occurring at 50 °C and 150 °C.

The remainder is either more tightly bound or less able to diffuse to the surface, and continues to evolve gradually up to about 600 °C.

Between 250 and 550 °C, an endothermic reaction occurring on the material, caused by the dihydroxylation of the octahedral sheet (constitutive water) of muscovite. On the TG curve, the hydroxyl groups of the tetrahedral sheet of muscovite are gradually removed up to 850 °C. An exothermic peak resulting by the crystal reformation (zeolite phase), then appears between 550 and 750 °C. It can also be inferred that the exothermic peaks at 850–1150 °C are due to the formation of nepheline (NaAlSiO₄).

A large and intense exothermic peak started from 1150 °C attributed to the structural reorganization.

3.2.4 Electrical Conductivity of Na-Sialate Geopolymer

As mentioned above, from 300 to 900 °C, Na-sialate geopolymer presents the same disordered structure of the nepheline. The crystal structure of Nepheline is characterized by layers of six-membered tetrahedral rings of exclusively oval
The rings are built up by regularly alternating AlO₄ and SiO₄ tetrahedral. The stacking of the layer’s parallel to the c axis results in a three-dimensional network containing channels that are occupied by the Na cations [38]. This topology is in favor of an easy movement of Na⁺ ions throughout the structure. For this reason, ionic migration in nepheline is extensively reported [39–41].

The electrical measurements were performed on geopolymer pellets sintered at 300 °C to remove free water. Figure 9 shows the impedance plot at different temperatures of the Na-sialate geopolymer. The data was refined with an electrical equivalent model, the polarization part of the sample and the electrode polarization were modeled by a constant phase element (CPE), while the migration part was represented by a resistance R. The complex impedance spectra were characterized by the appearance of a semi-circle centered below the x-axis. At room temperature, the diagram shows two contributions, with two tangled semicircles. These two relaxations, modeled by two parallel R//CPE circuits, correspond to ion migration in the grains and in the grain boundaries. The refinement of Na-Sialate geopolymer at room temperature gives bulk high ionic conductivity of about 5 × 10⁻⁵ S cm⁻¹. At higher temperatures, the characteristic frequencies become so close that it is impossible to distinguish the contributions. A total resistance comprising both grain and grain boundaries contribution is then determined.

According to these results, the temperature dependence of electrical conductivity can be analyzed following an Arrhenius plot (Fig. 10).

Showing Fig. 10, two different behaviors of the geopolymer can be evidenced. At low temperature, the total conductivity slowly decreases under 200 °C. This phenomenon can be attributed to the loss of water which was absorbed on the surface of the pellet leading to protonic conduction. Then, from 200 to 725 °C, the conduction properties seem to approximately follow an Arrhenius law both on heating and cooling with deduced activation energies which are 0.26 eV and 0.28 eV respectively.

Thus, Na-Sialate geopolymer presents lower σ than the specific ionic conductivity of single crystals of Na-Nepheline along the crystallographic c-axis [38] but higher than the value measured in the polycrystalline sample [16, 40, 41].

**4 Conclusions**

A new Na geopolymer was synthesized by mechanically mixing distilled water with a solution of sodium hydroxide and metakaolin with a ratio of Si/Al = 1 and Na/Al = 1, Al₂O₃ 2SiO₂ metakaolin was obtained from well crystallized commercial kaolin (clay).

The clay was a mixture of Kaolinite and Muscovite.

This geopolymer has been studied by X-ray powder diffraction, IR spectroscopy, thermal analysis and impedance spectroscopy dielectric.

It has been proven that the geopolymer has an amorphous character at room temperature and contains peaks related to the existence of muscovite nanoparticles.

During calcination, the change in cell volume of the different phases indicates the appearance of the nanocrystals relating to zeolite and muscovite between 300 °C and 600 °C.

Increasing the calcination temperature up to 950 allows us to obtain a crystalline material which is Nepheline (NaAlSiO₄).

The DTA thermogram shows that below 250 °C, that the sample exhibits about 14% mass loss linked to the evaporation of free water (~70%).
Between 250 and 550 °C, an endothermic reaction occurs on the material, caused by the dihydroxylation of the octahedral layer (consequent water) of the muscovite. These hydroxyl groups of the tetrahedral sheet of muscovite are gradually eliminated up to 850 °C. An exothermic peak resulting from crystal reformation (zeolite phase) then appears between 550 and 750 °C. It can also be deduced that the exothermic peaks at 850–1150 °C are due to the formation of nepheline (NaAlSiO₄).

A broad and intense exothermic peak started from 1150 °C attributed to the structural reorganization. The crystal structure of nepheline is characterized by layers of six-membered tetrahedral rings of exclusively oval conformation. The rings are formed by regularly alternating AlO₄ and SiO₄ tetrahedra. The stacking of layers parallel to the c axis gives a three-dimensional network containing channels occupied by Na⁺ cations. This topology favors easy movement of Na⁺ ions throughout the structure. For this reason, ionic migration in nepheline is widely reported [39–41].

The electrical measurements were carried out on geopolymer pellets sintered at 300 °C to eliminate free water. Refinement of the Na-Sialate geopolymer at room temperature yields a high mass ionic conductivity of about 5 × 10⁻⁵ S cm⁻¹. At higher temperatures, the characteristic frequencies become so close that it is impossible to distinguish the contributions. A total resistance including both the grain and grain boundary contribution is then determined.

At low temperature, the total conductivity decreases slowly below 200 °C; which can be attributed to the loss of water which has been absorbed on the surface of the pellet resulting in proton conduction. Then, from 200 to 725 °C, the conduction properties appear to follow approximately an Arrhenius law on both heating and cooling with inferred activation energies which are 0.26 eV and 0 respectively, 28 eV.

Thus, the Na-Sialate geopolymer exhibits a σ lower than the specific ionic conductivity of Na-Nepheline single crystals along the crystallographic c-axis [38] but higher than the value measured in the polycrystalline sample. As next research studies we will do the SEM and the TEM whose purpose is to study the morphology of geopolymer according to temperature as well as the sizes of the nanocrystallites.

Acknowledgements The authors would like to thank the Professor Mohamed Touni, for the beneficial discussions, and staff of ‘Le Mans Institute of Molecules and Materials (IMMM), CNRS UMR 6283, Le Mans University, avenue Olivier Messiaen, F-72085 LE MANS Cedex 9, France’

Funding The authors have not disclosed any funding.

Declarations

Conflict of interest The authors have not disclosed any competing interests.

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