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Title

Combined experimental and computational prediction of the piezoresistivity of alkali activated inorganic polymers

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KEYWORDS

Piezoresistance, electron mobility, inorganic polymer, alkali activated materials, Kubo-greenwood theorem.

ABSTRACT

The incorporation of smart building materials into construction will improve the working life of structures and infrastructure around the globe. Unfortunately, conventional smart building materials are cost prohibitive because of the self-sensing additives required. Alkali activated inorganic polymers are a promising low-cost and environmentally friendly alternative that exhibit intrinsic self-sensing properties, without need for self-sensing additives. An improved methodology has been developed to quantify the self-sensing piezoresistivity of these materials. Experimental measurements reveal a strong intrinsic piezoresistivity, up to 12% at 10 MPa. The results agree with a first-principles model of the theoretical piezoresistivity of an alkali activated inorganic polymer from quantum mechanical perturbation theory. This first-of-its-kind computation provides a mechanistic explanation for the origin of intrinsic piezoresistivity in inorganic polymers.

1 Introduction

Smart building materials are multifunctional construction materials designed to improve the maintenance of structures and prolong the working life of buildings.[1] They are typically high mechanical strength materials that have been modified with additives to impart self-healing or self-sensing properties. Self-sensing is an intelligent material property wherein the material supports a nonintrusive detection method that allows continuous assessment of the material's mechanical state. Self-sensing building materials can detect pressure, vibrations, and abnormal strain in a structural member. This has led to their proposal for a wide variety of applications including structural health monitoring, corrosion detection, seismic damage detection, traffic sensing, and border security monitoring.[2–5] However, the additives used to impart self-sensing properties severely limit the affordability and the durability of the materials.[6–8] In this work, alkali activated inorganic polymers are demonstrated to exhibit self-sensing behavior intrinsically, circumventing the drawbacks of self-sensing additives while maintaining high mechanical strength. To understand the origin of this unusual property, the electrical conductivity of inorganic polymers was modeled from first principles by quantum mechanical perturbation theory.

Conventional self-sensing building materials are composites of an inorganic matrix material, most commonly Portland cement, and a conductive additive, typically carbon fiber.[1] The self-sensing property is achieved through the piezoresistivity of the composite, in which the electrical conductivity of the composite is sensitive to strain-induced changes in the tunneling distance between the conducting carbon fibers. This methodology requires a high content of carbon fiber (e.g. 15%) and is sensitive to the dispersion of the fibers and changes in the fiber-matrix

interface, which has drawn questions about its affordability, reproducibility, and longevity of composites as self-sensing building materials.[9,10] An ideal improvement would be the identification of a building material that is self-sensing intrinsically, without the need for conductive additives. In one recent study, a potassium activated inorganic polymer was shown to exhibit this property, but the extent of its presence in other alkali activated inorganic polymers and the mechanisms behind the intrinsic piezoresistivity are unknown.[11]

Alkali activated materials are amorphous inorganic polymers with ceramic-like properties, formed through a polycondensation reaction of aluminum and silicon oxides dissolved in the presence of a high pH activator.[12] Their elemental composition is analogous to zeolites, but they are formed at room temperature which has attracted many comparisons to Portland cement.[13] Generally, they exhibit high mechanical properties and are considered a low-cost and environmentally friendly alternative to Portland cement because they can be synthesized from industrial byproducts that are rich in aluminum and silicon oxides, such as blast furnace slag and coal fly ash.[14] Alkali activated inorganic polymers have been investigated as a matrix material for self-sensing composites with conductive additives.[15–18] However, their intrinsic piezoresistivity was found to be negligible with conventional measurement methods.

Conventionally, piezoresistivity is measured under direct current conditions with cyclic loading of compressive stress. Cyclic loading simulates how a self-sensing material may behave in applications with transient loads, such as in traffic monitoring.[5] This has created a challenge to the deployment of self-sensing building materials in other applications of interest, such as structural health monitoring and corrosion detection, in which loading changes are static. Despite interest in these applications, few studies have measured piezoresistivity under static conditions

and it has been observed that some piezoresistive materials are only self-sensing under cyclic loading.[10,19]

This investigation seeks to develop a new understanding of the intrinsic piezoresistivity of alkali activated inorganic polymers. Unlike for conventional self-sensing composites, the mechanisms behind intrinsic piezoresistivity are not known. Modeling of the theoretical properties of inorganic polymers only recently became possible due to molecular dynamics simulations of the complex amorphous structure.[20] The goal of this study is to, for the first time in the scientific literature, use a structural model of an inorganic polymer to compute a theoretical property. The piezoresistivity is calculated by applying the Kubo-Greenwood theorem, a first-principles quantum mechanical expression for electrical conductivity. The Kubo-Greenwood theorem has never previously been applied to a complex amorphous material and the results are corroborated by experimental measurements. An improved experimental methodology has been developed to accurately measure the intrinsic piezoresistivity of inorganic polymers under static loading conditions. The results present inorganic polymers as an environmentally friendly alternative for smart building materials without the drawbacks of conventional composites.

2 Experimental methods

Experimental specimens of inorganic polymer were prepared through the alkali activation of fly ash (ProAsh®, Separation Technologies Inc.). The specimens were prepared in triplicate using sodium metasilicate and silica fume to achieve an elemental ratio of 1.1:1:3 Na/Al/Si (sodium/aluminum/silicon). A water/binder ratio of 0.25 was used to form a flowable paste. The mixture was prepared in a low-shear mixer and cast into stainless steel molds which were sealed

to prevent moisture loss during curing. Specimen for mechanical properties were prepared and tested in accordance with ASTM C109 and ASTM C348.[21,22] For piezoresistivity specimens, 5 cm cube molds were modified with a bracket to maintain two steel mesh electrodes at a spacing of 1 cm apart. The specimens were cured in a saturated humidity chamber at 60ºC for 7 days to ensure complete reaction. Prior to piezoresistivity measurements, excess moisture was gently removed without damaging the native hydrates by drying at 60ºC for 7 days in a dry oven. For comparison, a piezoresistivity specimen of Portland cement was prepared with a water/binder ratio of 0.3 at cured at room temperature for 28 days prior to testing.

The piezoresistivity was calculated by measuring the conductivity of the specimens with an alternating current source, adapted from the method described by *Saafi et al*.[11] The electrical properties were measured using the experimental setup shown in Figure 1(c). Compressive stress was applied at 1 MPa intervals up to 10 MPa in a direction orthogonal to the orientation of the electrodes. At each interval, the conductivity was measured using a Wheatstone bridge containing resistors of 860 kΩ and a 10-volt alternating current source between 10 Hz and 10 MHz. The resistance of the bridge resistors was selected to be close to the expected resistance of the specimen, based on published values for other alkali activated fly ash inorganic polymers, to maximize the accuracy of the measurement.[23] The source voltage, V_{source}, and the voltage across the electrodes, $V_{electrodes}$, were measured using an InfiniiVision® 3034A oscilloscope (Agilent Technologies Inc.) and the sample resistance, R_{sample}, was calculated using the following equation, derived from Ohms law:

$$R_{sample} = R_3 \frac{R_2 - \frac{V_{electrodes}}{V_{source}}(R_1 + R_2)}{R_1 + \frac{V_{electrodes}}{V_{source}}(R_1 + R_2)}$$

where R_1, R_2, and R_3 are the resistances of the of the bridge resistors in the configuration shown in Figure 1(c). The self-sensing piezoresistivity was quantified, by convention, as the fractional change in resistance (FCR) defined by:

$$FCR\ (\sigma, \varepsilon) = \frac{\rho_o - \rho_{\sigma,\varepsilon}}{\rho_o}$$

where ρ_o is the unstrained resistivity, and $\rho_{\sigma,\varepsilon}$ is the resistivity at stress σ or strain ε, respectively.

3 Theoretical calculations

A structural model of an alkali activated inorganic polymer was generated from the molecular dynamics simulation presented by *Zhang et al.*[20] A 240 atom structure, representing at 1.5 nm cube of inorganic polymer, was modeled in the opensource software Quantum Espresso. The structure achieved the elemental ratio of 1.1:1:3 Na/Al/Si and an H_2O/Al_2O_3 ratio of 10.75. The structure was relaxed using the Broyden-Fletcher-Goldfarb-Shanno algorithm to minimize the free energy as described by *Fletcher*.[24] Following the methodology explained by *Calderin et al.,*[25] Quantum Espresso was used to compute the theoretical electrical conductivity of this structure and a projector augmented wave pseudopotential was assumed for each atom. The self-consistent field calculations were conducted to establish boundary conditions. This calculation was solved iteratively for the kinetic energy (Wfc) and potential energy (Rho) cut-offs in ranges of 40-95 and 300-800, respectively, using 111 kpoint and David diagonalization for each element present in the inorganic polymer. The self-consistent field parameters are given in Table 1 and were used in Quantum Espresso to solve the Kubo-Greenwood theorem of electrical conductivity.

Table 1: Self-consistent field calculation results for the alkali activated inorganic polymer.

Element	Wfc	Rho
Sodium	85	750
Aluminum	75	450
Silicon	85	750
Hydrogen	65	700
Oxygen	45	600

The Kubo-Greenwood theorem is a quantum mechanical expression that describes the transport coefficient of electrons through a structure for a perturbation of frequency ω.[26,27] In the general case, the electrical conductivity, σ, is given by the expression:

$$\sigma(\omega) = \frac{2ie^2\hbar^3}{m_e^2 V} \sum_{m,m'} \frac{\Delta f_{m'm}}{\Delta \epsilon_{mm'}} \frac{\langle m|\nabla|m'\rangle \langle m'|\nabla|m\rangle}{(\Delta \epsilon_{mm'} - \hbar\omega + i\delta/2)}$$

for each pair of atoms, m and m', where f is the Fermi-Dirac occupation number, ϵ is the corresponding eigenvalue, and V is the unit cell volume. The theorem was solved with the given parameters and structure using the methodology described by *Calderin et al.*[25] Interested readers are directed towards that manuscript for an in-depth explanation. Notably, the application of the Kubo-Greenwood theorem yields the theoretical conductivity of the material, under the assumption of no atomic movement and no interfacial contributions.[28] The theoretical self-sensing behavior of the inorganic polymer was calculated by applying artificial strain to the structural model. The structure was strained at intervals up to 0.8%. At each interval, the Kubo-Greenwood theorem was applied to yield a new strained conductivity and the piezoresistivity was quantified as fractional change in resistance (FCR).

4 Results and discussion

In this study, a novel method is demonstrated to measure the piezoresistivity of smart building materials. The method is adapted from the work presented by *Saafi et al*., which demonstrated intrinsic piezoresistivity in an inorganic polymers using alternating current conditions.[11] Alternating current eliminates the effect of polarization in insulating materials, such as inorganic polymers, which causes the conductivity to decline over time during the measurement.[10] Inorganic polymers have high polarizability making them particularly susceptible this form of systemic error.[23] Polarization has lead other investigators to conclude that piezoresistivity in inorganic polymers can only be observed under cyclic loading conditions, which is impractical for most applications of in-situ self-sensing.[19] However, it has been recently demonstrated that piezoresistive composites can be used under static loading conditions with alternating current.[18]

This study demonstrates that this methodology can be used to measure the intrinsic piezoresistivity of inorganic polymers under static loading conditions. The electrical resistance was measured with an alternating current source using a Wheatstone bridge and oscilloscope, as shown in Figure 1(c). At a testing frequency of 10 Hz, the measurements could be conducted rapidly and reliably, leading to minimal standard deviation, depicted as error bars in the figures. This methodology was used to assess the piezoresistivity of the alkali activated inorganic polymer and Portland cement. The piezoresistivity was quantified as the fractional change in resistance (FCR) at each compressive stress interval in Figure 1(a). The inorganic polymer exhibited strong piezoresistivity, up to 12.2% FCR at 10 MPa. The FCR closely follows a logarithmic trend (R^2>0.98) with a stress sensitivity coefficient of 0.05 MPa^{-1}.

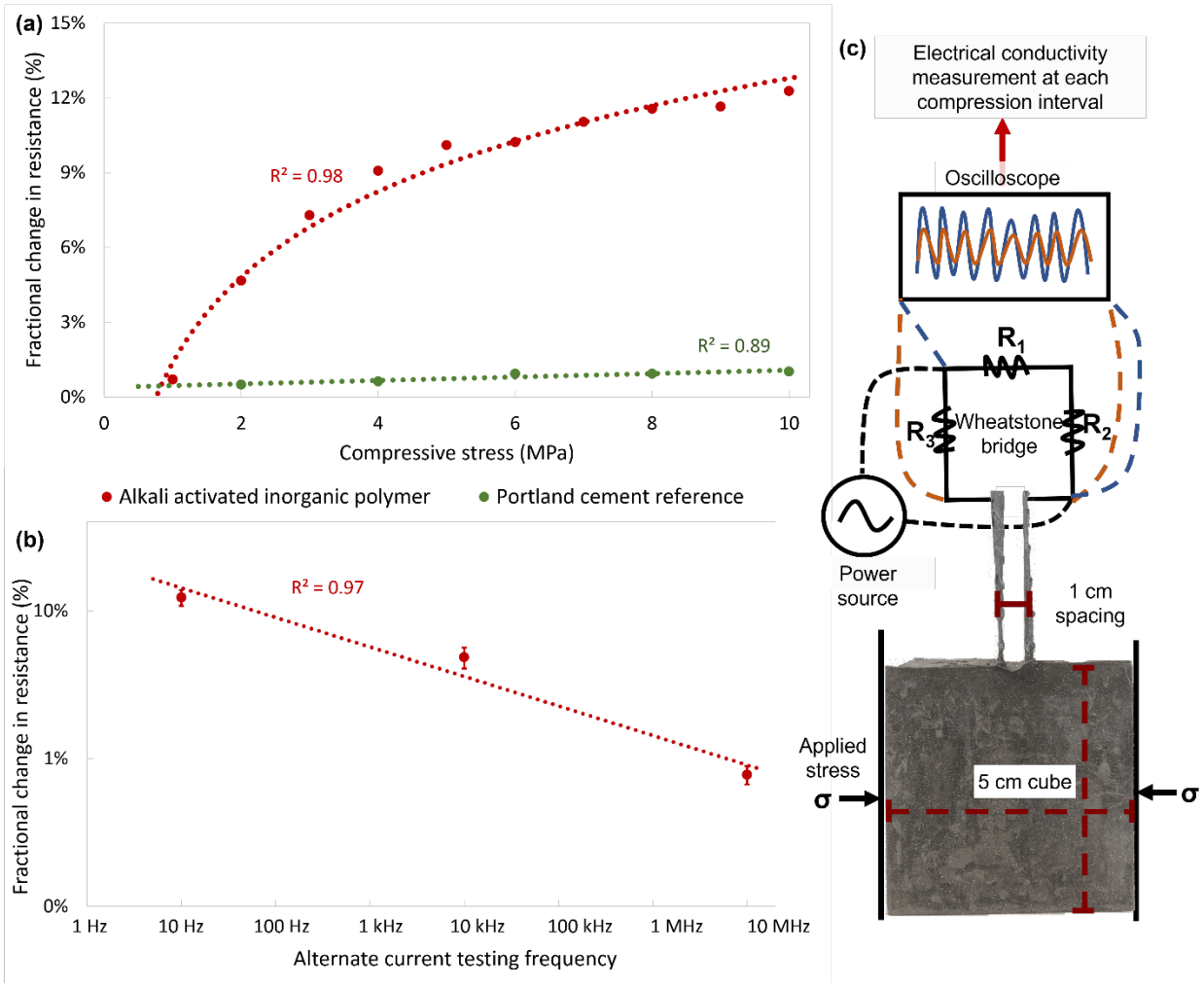

Figure 1. (a) The piezoresistivity of alkali activated inorganic polymer and Portland cement at 10 Hz, (b) the frequency dependence of the piezoresistivity of inorganic polymers at 10 MPa, and (c) the experimental method for the piezoresistivity measurement.

The piezoresistivity of the inorganic polymer was significantly higher than prior studies of inorganic polymers under cyclic loading. Inorganic polymers without conductive additives have FCR below 1% when measured with direct current under a cyclic loading.[16,17] The use of alternating current increases the detectable FCR by a factor of 10, opening the door to future use of inorganic polymers as self-sensing materials. Portland cement is known to have negligible

FCR under cyclic loading conditions.[10] However, Figure 1(a) reveals that alternating current testing does not bolster the FCR of Portland cement and, thus, piezoresistivity cannot be achieved without conductive additives. This indicates that while the intrinsic piezoresistivity of inorganic polymers can be obscured by polarization, but it remains a unique property of inorganic polymers and an advantage of inorganic polymers over Portland cement. At 12.2% FCR, the inorganic polymer has intrinsic piezoresistivity that rivals piezoresistive composites. The results are in the range of the 16.5% FCR of inorganic polymer composites containing graphene and the 7% FCR of carbon fiber composites at 10 MPa.[15,18] The intrinsic FCR of inorganic polymers is comparable to the 13-20% FCR that has been achieved with cement composites containing carbon fiber and carbon nanotubes at 10 MPa.[9,29] Inorganic polymers are superior because they reach similar FCR intrinsically, without the added cost, environmental impact, or durability reduction caused by carbon fiber addition.

The alternating current measurement method was effective for measuring the piezoresistivity but was found to be sensitive to the power source frequency. This is a consequence of the frequency dependence of the conductivity of insulators, including alkali activated materials, which has been reported to vary over orders of magnitude across large ranges of frequency.[23] The piezoresistivity of the inorganic polymers was found to closely follow a power function (R^2>0.97) with frequency, as shown in Figure 1(b). The piezoresistivity is maximized at low frequency. This can be understood by considering the piezoresistivity mechanisms that are active in the material, which are discussed below. 10 Hz was selected as a compromise of rapid measurement speed, to avoid fluctuations in the compressive loading of the specimen, and the high piezoresistive response of the material. Future studies that employ this methodology must be consistent with the measurement frequency to make faithful comparisons.

Understanding the piezoresistive mechanisms within inorganic polymers requires modeling electrical changes in the structure during strain. Prior studies have suggested, without evidence, that the strong piezoresistive response in inorganic polymers originates from the ionic conductivity of alkali cations through the amorphous structure.[11,30] This study seeks to challenge that conjecture and prove that the piezoresistivity arises, at least in part, from changes in electron mobility. To accomplish this goal, the electrical properties were modeled from first principles using the Kubo-Greenwood theorem. This is a first-of-its-kind computation of the theoretical conductivity of an amorphous ceramic-like material. The computation does not consider the conduction or movement of ions, assuming fixed atomic positions.[28] This will elucidate the electron mobility contribution to the piezoresistivity as artificial strains are applied to the structure.

The modeled alkali activated inorganic polymer has the same elemental ratio (1.1:1:3 Na/Al/Si) as the experimental specimens that were tested. The relaxed model is shown in Figure 2(a). The piezoresistivity, quantified as the fractional change in resistance (FCR), was computed at four intervals of strain, up to 0.8%. The piezoresistivity is shown in Figure 2(b) to increase linearly (R^2>0.99) up to 14% FCR with a strain sensitivity coefficient of 17.8. This definitively proves that inorganic polymers exhibit piezoresistivity as an intrinsic material property, not caused by an undocumented phase or other phenomena.

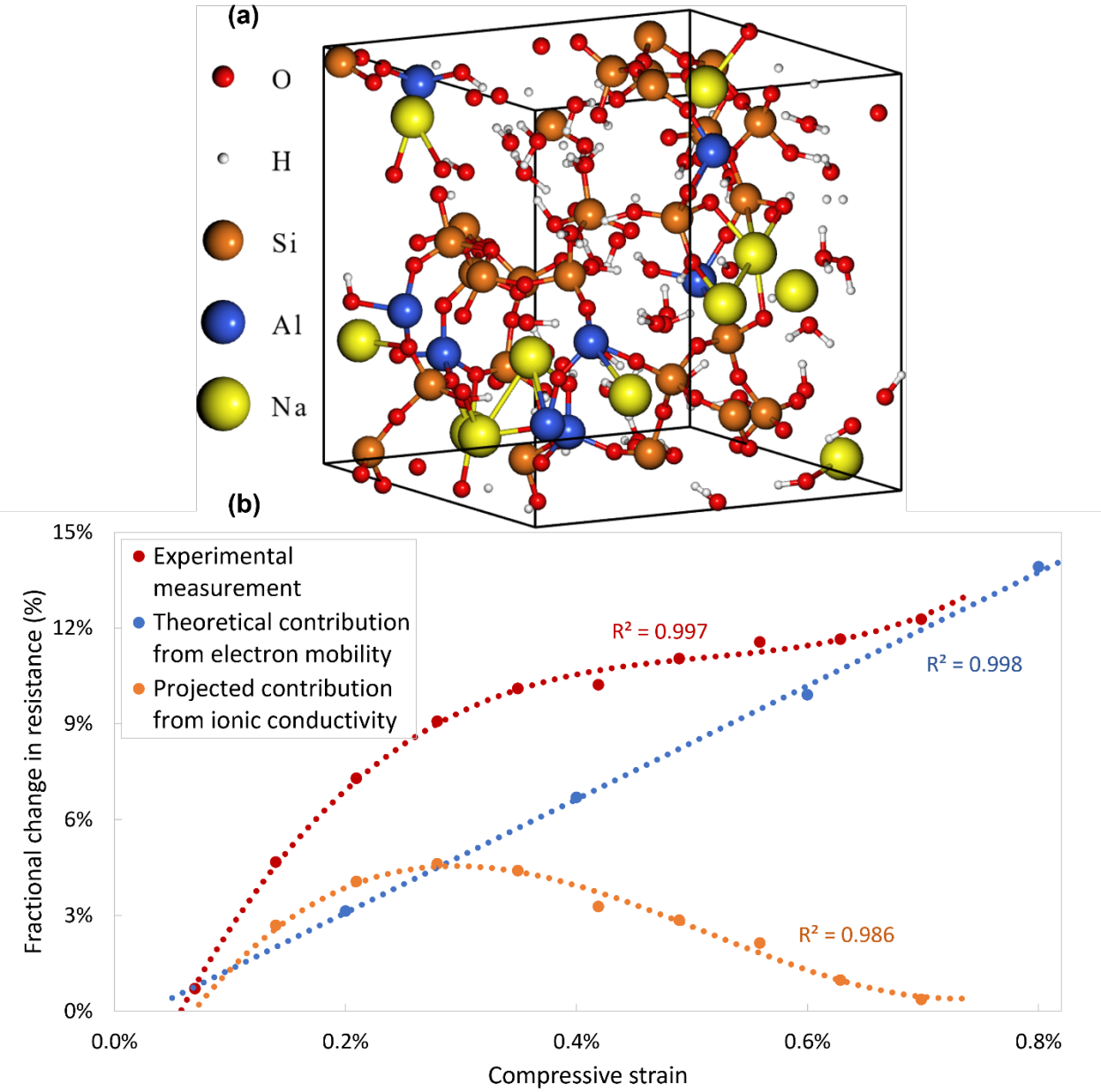

Figure 2. (a) The structural model of the alkali activated inorganic polymer of elemental ratio 1.1:1:3 Na/Al/Si after relaxation in Quantum Espresso and (b) the theoretical piezoresistivity of the inorganic polymer and the contribution from the ionic conductivity.

The result also proves that changes in electron mobility are a major contributor to the piezoresistive response. However, the electronic piezoresistivity is lower than what was observed

experimentally, particularly at low strains. The difference between the theoretical calculation and the experimental results can be attributed to the ionic conductivity of the alkali activated inorganic polymer. The contribution of the ionic conductivity, as shown in Figure 2(b), is significant at small strains but declines at high strain values. Charge hopping mechanisms for ionic mobility are enhanced when strain is applied because the distance between neighboring sites is reduced.[30] However, at higher strains, the conduction pathways are constricted, resulting in lower ionic mobility. The ionic contribution becomes negligible at higher strains and the experimentally measured FCR converges with the predicted piezoresistivity due to the electronic mobility. The two factors together result in the nonlinear piezoresistive behavior that is observed experimentally.

Smart building materials are required to exhibit high mechanical properties, excellent durability, and useful self-sensing properties. Alkali activated inorganic polymers have been extensively studied in the literature as a cement alternative with greater mechanical properties and durability than Portland cement.[31] The compressive and flexural strengths of the experimental specimen investigated in this work were measured to be 31.9 and 3.90 MPa, respectively. This meets the requirements for infrastructure applications and is consistent with typical values for alkali activated fly ashes.[32] With high strength and intrinsic piezoresistivity, this inorganic polymer is a competitive alternative to cement composites proposed for smart building materials. The use of alkali activated materials as piezoresistive building materials circumvents the challenges facing conventional cement-carbon fiber composites including high cost, dispersion reproducibility, and limited durability.[6,7,10] Alkali activated inorganic polymers are a superior smart building material. The measurement method and theoretical model for the intrinsic piezoresistivity presented in this work create a framework for future investigations into this novel material

solution. Future studies should apply compositional modifications to the inorganic polymer to bolster the mechanical properties and investigate the impact of these compositional changes on the piezoresistive behavior.

5 Conclusion

Alkali activated inorganic polymers are a superior smart building material with high mechanical strength and intrinsic self-sensing behavior. However, to date, they have been overlooked because their structure and properties are not well understood. In this study, a combined theoretical and experimental approach has been applied to elucidate the nature of the intrinsic piezoresistivity of alkali activated inorganic polymers. The results shed light on the origin of this property in inorganic polymers and demonstrate a methodology that can be used to investigate the theoretical properties of other smart building materials. Alkali activated inorganic polymers offer a superior combination of strength and self-sensing behavior compared to conventional cement composites, without the need for self-sensing additives.

6 Data and software availability

The atomic positions generated for the model and used in the calculations are available in the supplemental information. The computations were completed with Quantum Espresso, an open-source software.

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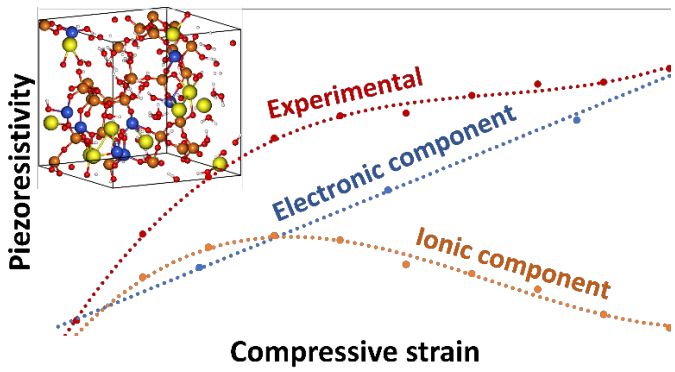
TOC Graphic


Piezoresistivity
Experimental
Electronic component
Ionic component
Compressive strain