



## Research article

## Towards the development of stable and efficient novel waste ceramics composites



Mohammad Sohail<sup>a,b,\*</sup>, Sanaullah Khan<sup>d</sup>, M. Saleem Khan<sup>b</sup>, Ihsan Ullah<sup>a</sup>, Muhammad Omer<sup>a</sup>, Noor Saeed<sup>c</sup>, Sabiha Sultana<sup>c</sup>, Adnan Adnan<sup>a</sup>, Adnan Shahzad<sup>a</sup>, Mian Gul Sayyed<sup>a</sup>

<sup>a</sup> Institute of Chemical Sciences, University of Swat, KP, Pakistan

<sup>b</sup> National Center of Excellence in Physical Chemistry, University of Peshawar, KP, Pakistan

<sup>c</sup> Islamia College University Peshawar, KP, Pakistan

<sup>d</sup> Department of Chemistry, Swabi Women University, KP, Pakistan

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## ABSTRACT

Natural resources are non-renewable and facing a regular depletion due to their immense use which demands new and additional material's reserves, recycling technologies and materials with no or less bad environmental effects. Reuse of waste materials will be rewarding technically, economically and environmentally. Here, we report the incorporation of industrial ceramic wastes in polymer matrix as composite materials to investigate their potentials for various applications. Ceramic wastes were collected from the premises of ceramic producing industries located at Peshawar (Pakistan). The composites of ceramic particles and polyaniline (PANI) were produced via *in-situ* free polymerization technique. SEM and FT-IR analysis confirmed composite formation. Thermal, dielectric and mechanical properties of the prepared materials were studied. It was found that both the constituent materials (ceramic and polymer) have a synergistic effect on each other. At one hand, ceramic wastes support and enhance the thermal and mechanical properties of the polymer in composites and the polymer in turn beautify the wastes with good dielectric and electrical properties. Based on their properties, the low cost and environmentally friendly novel composites could be used for various applications such as semi-conductors, capacitors and microwave devices.

## 1. Introduction

Reliable, cheap, clean, sustainable energy and green environment has been a keystone of the world's increasing economic growth and prosperity since the beginning of the industrial revolt. Stress on the scope and importance of sustainability and reprocessing has become increasingly known and interpreted by the academia and industry over the last three decades. Local ceramic industries mainly generate ceramic bricks, roof tiles, and floor for commercial purposes. About 2–3% of the products are rejected during grinding, pressing and plating processes. The refused materials (pellets, large particulates and powders) cannot be recycled within the industrial plant which are discarded nearby in the open ditches which causes serious environmental issues. Natural resources are declining increasingly due to their abandoned use. Recycling of these ceramic wastes suggest, energy saving, cost reduction, alternative products, prohibit landfilling and reduce reliance on natural resources. Ceramic wastes have been found to have a tough texture, good

mechanical and thermal stability and mostly unaffected by chemical and biological degradation [1]. A lot of work has been done to use the waste materials for better purposes since last two decades. Ceramic waste has been utilized in the production of thermally stable geo-polymers [2]. Enhanced mechanical resistance was investigated in alumina waste based polymer composites [3]. PANI based red mud composites were prepared by Gok and his coworkers and examined conductivity in the range of 0.42–5.2 Scm<sup>-1</sup> [4]. Ceramic waste used in lime mortars showed effective mechanical strength for sustainable material [5]. Siqueria and coworkers observed about 100% flexural strength of waste ceramic composites [6]. The possible application of unblended ceramic waste has also been examined by several people, for example, ceramic sanitary ware [7], clay roof tiles [8] and ceramic bricks and rough fraction of ceramic waste [9].

While thermal stability, mechanical strength, heat and reagent resistance properties are concerned, ceramic materials are hard and durable. Ceramics are utilized in dental materials and porous media

\* Corresponding author.

E-mail address: [msohail2000@gmail.com](mailto:msohail2000@gmail.com) (M. Sohail).

**Table 1.** Density of the samples.

Sample codes	Density (g/cm <sup>3</sup> )
C1	2.52
C2	2.66
C3	2.67
C1P	1.87
C2P	1.52
C3P	1.16

combustion due to efficient mechanical/rheological properties [10]. Such properties enable ceramic materials to be used as fillers with polymers to get composites with enhanced assets for potent applications. Ceramic wastes may be a best option in this regard as they have almost the same characteristics as precursors with zero cost.

In the present study, we investigated ceramic wastes obtained from within and surroundings of three local roof tiles producing ceramic industries. Various environmental problems such as contamination, land occupation, water, soil and dust pollution are caused by these wastes. The soil of the area near these industrial plants is barren and unable to grow plantations due to surface water locking by the wastes. Recognition of silicosis is also expected in the workers near the industrial plant which is a serious health problem and needs adequate investigation. Use of the ceramic wastes in polymer composite as fillers is an environmentally comprehensive way to reduce their burden and pollution issues. Polyaniline (PANI) was used as matrix with the waste particle. Various physicochemical, dielectric and mechanical properties of the composite materials were studied to find the durability of the materials for possible technological applications.

## 2. Experimental

### 2.1. Collection and activation of ceramic wastes

Three ceramic roof tiles producing industries viz. Fort, Complex and Hussain tiles situated in the industrial zone Peshawar (Pakistan) were

visited to get the wastes. The three samples were properly washed subsequently with normal and distilled water to remove dust and soil particles. Afterwards, the samples were subjected to heat for drying and were ground to powder. The powders were passed via 220 $\mu$  mesh to get particles of the same dimensions. Fort Ceramics, Marble Complex and Hussain Marbles were named as C1, C2 and C3 respectively. EDX analysis was done to know about the chemical composition of the waste samples as shown in Table 1.

### 2.2. Preparation of waste ceramic composites

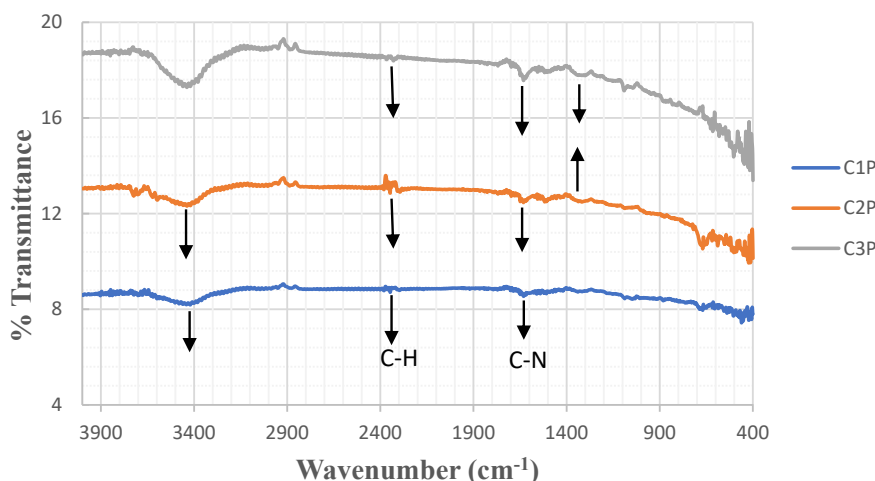
*In situ*, free-radical precipitation polymerization route was used for this purpose. During experiment, 250 mg of each C1, C2 and C3 waste was dissolved in 100 ml deionized water in three separate 500 ml beakers under magnetic stirring at 0–5 °C. Later, 10% aniline and 0.1M ammonium persulphate solutions were prepared in 1M HCl and an amount of 25 ml of each solution was put dropwise to all the beakers and were kept under stirring for 3 h. Afterward overnight digestion of the solutions was carried out. The solutions were centrifuged at 4000 rpm thrice to remove soluble impurities as supernatants. The residues were washed three times with water and were dried at 70 °C. The final products were named as C1P, C2P and C3P.

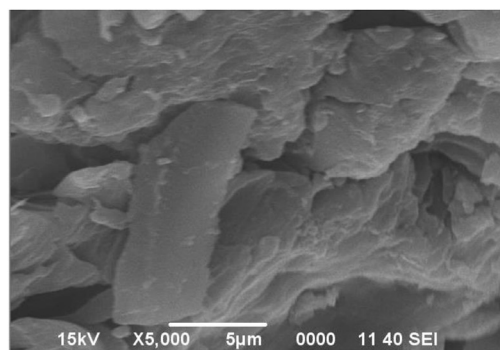
### 2.3. Characterization

Different techniques were used to analyze various properties of the prepared composites. The composite formation between ceramic wastes and PANI was confirmed via FT-IR spectroscopy (IR Model Prestige-21, USA). Micrographs of the composites were analyzed by JEOL Scanning Electron Microscope Model JSM-5910 (Japan). Thermal properties of the prepared materials were investigated by TGA (Diamond TG/DTA PerkinElmer, USA). Dielectric properties of the samples were examined with RF Impedance/Material Analyzer (Agilent E4 997 A, USA). Rheology of the composites was performed by Anton Paar Rheometer (Physica MCR 301 Germany).

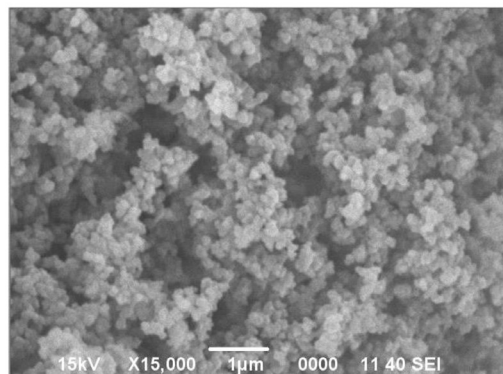
**Table 2.** Elemental composition of the collected ceramic wastes.

Waste Samples	Elemental composition Weight%									
	Si	AL	K	Ca	Fe	Ti	Mg	Na	C	O
C1	23.65	8.64	1.36	4.46	3.55	0.40	2.46	0.35	—	55.14
C2	—	—	—	37.94	—	—	—	—	10.15	51.92
C3	0.31	—	—	36.63	—	—	0.30	—	10.80	51.69

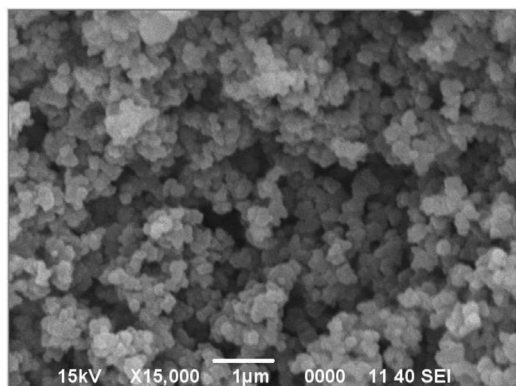
**Figure 1.** FT-IR spectra for functional group determination of the prepared composites.



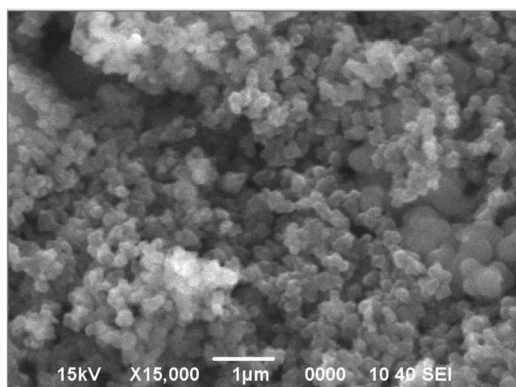
(a)



(b)

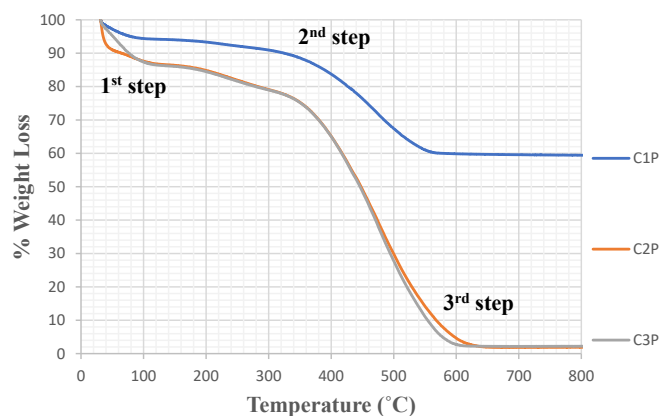


(c)



(d)

**Figure 2.** Micrographs representing the composite formation: (a) C1P, (b) C2P and (c) C3P.



**Figure 3.** Thermograms for C1P, C2P and C3P composite materials.

### 3. Results and discussion

#### 3.1. Density of the samples

Archimedes' principle-based density values were measured using a density meter as shown in Table 2. It is interesting to note that all the three composites display a lower density relative to their ceramic counterpart. This may be linked with the occurrence of cavities in the samples as shown in SEM micrographs. The low density of such composites is of great importance for application in the aerospace field due to the crucial demand of spacecraft for light materials [11].

#### 3.2. Structural analysis

Figure 1 shows the FT-IR transmission plots of two-phase C1P, C2P, and C3P composites. A broad peak at about  $3400\text{ cm}^{-1}$  is attributed to O–H stretching vibrations in two-phase composites due to  $\text{H}_2\text{O}$  molecules entrapped in the samples [4].  $900\text{--}1400\text{ cm}^{-1}$  bands represent C–H group in PANI benzonide rings [12]. At about  $1080\text{--}1089\text{ cm}^{-1}$  active C–H in-plane bending vibration appears. The C–C stretching in the aniline rings of PANI chain is shown by a peak at  $1230\text{ cm}^{-1}$  in the spectra represent. In C1,  $1058\text{ cm}^{-1}$  peak denotes Si–O vibration [4]. The short bands at  $400\text{--}900\text{ cm}^{-1}$  shows metal-oxygen interactions in the composites [13].

#### 3.3. Surface morphology

Figure 2a, b, c and d show the SEM micrographs of pure PANI, C1P, C2P and C3P respectively. Pure PANI shows sheets/large pellets like morphology. There are also present cavities and big holes in the polymer which provide a space for accommodating the incoming ceramic particles. It is obvious that the materials exhibit micro-clusters and porous morphologies. Ceramic particles deliver self-assembled phase over PANI chains which behave like broccoli containing several small flower buds. The presence of microspores is assumed to be due to the grafting of ceramic particles within the PANI network [14]. The polymer surface is

**Table 3.** Stability range and residues left during TGA analysis of the composites.

Samples	Stability Range		Residues
	$T_{di}$ (C)	$T_{max}$ (C)	$Y_c$ at $800\text{ }^\circ\text{C}$ (Weight %)
C1P	340	560	59.45
C2P	358	620	2.21
C3P	358	615	2.00

seen to be covered by the dispersed spherical waste ceramic particles. It is expected that the particles are not only distributed over the PANI surface but also got an access into the interior of the PANI sheets as reported for red mud/PANI composites [4]. The holes present in the polymer are of enough size to accommodate the initial incoming ceramic particles while the latter incoming particles set over the surface of polymer and hence producing cauliflower like morphology. Ceramic particles show size in micro-dimensions and due to the unavailability of resources actual size of the particles was not confirmed. The presence of embedded large aggregates over the surfaces may be due the presence of water vapors that have compacted the particles. PANI chains overlapping might have occurred during the drying that results in agglomeration as reported in previous literature [15]. In agreement with the density calculations (Table 2), residual porosity observed in composites suggests their application in thermal insulation.

### 3.4. Thermal properties

TGA (Figure 3) [1] confirmed that the decomposition of composite materials completes in three steps:

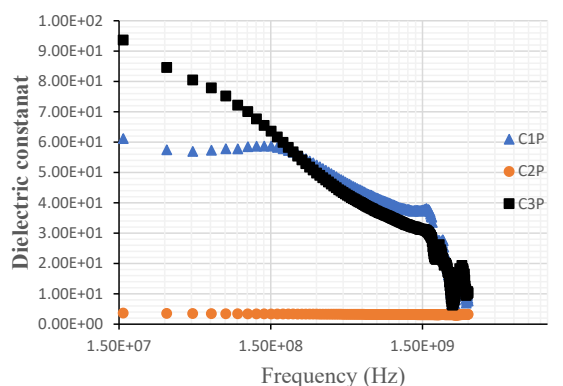
- I. First step at around 70–90 °C with a weight loss of 6% for C1P, 8% for C2P and 10% for C3P is due to the loss of water molecules.
- II. The second step (340–360 °C) shows the initial decomposition temperature ( $T_{di}$ ) while at 560–615 °C represents temperature of maximum degradation ( $T_{max}$ ) for the composites.
- III. The last step beyond 615 °C shows the complete degradation of PANI chains in the composites which leads to the production of gases [16]. The residues left at 800 °C is the char (PANI) and diffused form of ceramic wastes.

The ceramic wastes are thermally stable (up to 700 °C) as reported earlier (Khan et al., 2016). Similarly, PANI have thermal stability in the range from 200–400 °C as reported in literature [17, 18]. The insertion of ceramic wastes in PANI chains has enhanced the thermal stability of the polymer. The enhanced stability of conducting PANI in composites makes it of vast significance for high temperature applications. Table 3 shows  $T_{di}$  and  $T_{max}$  for C1P, C2P and C3P which specifies that they have about similar degradation mechanism.

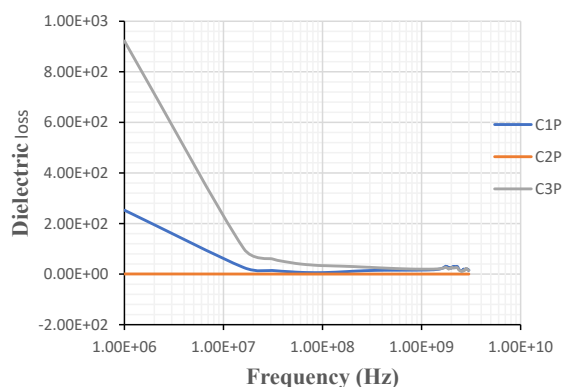
### 3.5. Dielectric properties

Dielectric properties such as dielectric constant ( $\epsilon'$ ), dielectric loss ( $\epsilon''$ ) and loss tangent ( $\tan \delta$ ) of the composites were analyzed (Figure 4a, b and c). It is obvious that the three composites show high range of dielectric properties at low current frequency range. Maxwell Wegner theory [19] confirmation is valid at low range. The dielectric material exhibits interfacial polarization at low frequency thus improves the dielectric behavior of the material. With increasing frequency, the dielectric properties of the materials are decreased. This is due to the inability of charge carriers to line up and adjust themselves with the alternating frequency and as a result relaxation occurs. Resonance occurs at high frequency (2 GHz) which results in *non-Debye* effect because of the occurrence of equilibrium in between the frequencies of electron hopping and that of applied field. It is accredited to the absence of coordination between charge carriers at high frequency due to which the dipoles present in the composites are unable to place themselves properly [20]. Typically,  $\epsilon''$  value designates the conductivity ( $\sigma$ ) of a material (high  $\epsilon'' \sim$  high  $\sigma$ ). In this case, C3P ( $\epsilon'' = 129$  units) have the highest conductivity followed by C1P ( $\epsilon'' = 60$  units).

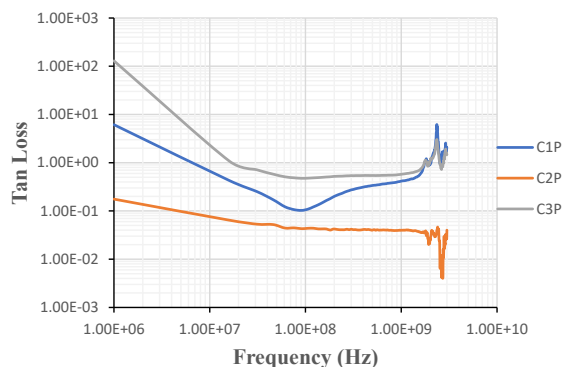
Frequency dependent *ac* conductivity ( $\sigma_{ac}$ ) of the composites was calculated by the following equation:



(a)



(b)



(c)

Figure 4. Dielectric behavior of composites: (a) Dielectric constant, (b) Dielectric loss, (c) Tan Loss.

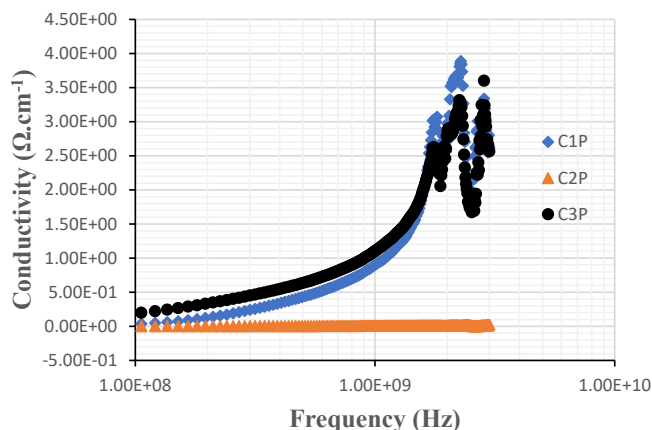


Figure 5. Frequency dependent *Ac* conductivity of the prepared composites.

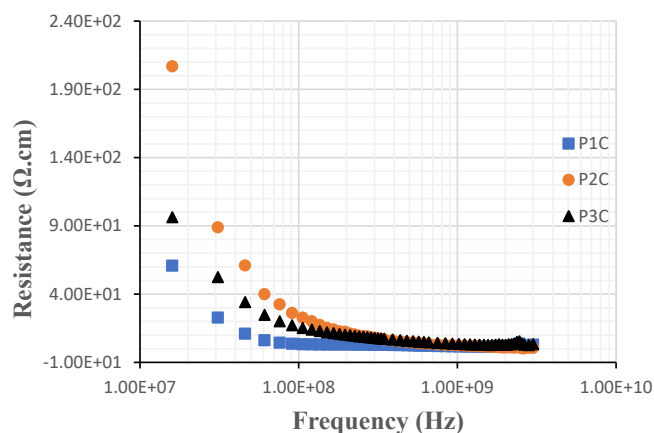


Figure 6. Decrease in resistivity with increasing current frequency.

$$\sigma_{ac} = \varepsilon' \varepsilon'' \omega \tan \delta \quad (1)$$

where  $\omega$  is the angular frequency of the applied field ( $\omega = 2\pi f$ ) and  $\varepsilon$  is the permittivity of free space ( $8.85 \times 10^{-12} \text{ F m}^{-1}$ ). Conductivity is increasing with increasing frequency for C1P and C3P while C2P is not so much active in this regard just like its other dielectric properties as shown in Figure 5. High conductivity shows the reorientation of dipoles with the electric field which is assumed to be due to the occurrence of Maxwell-Wagner interfacial polarization at high frequency. The charge carriers accumulate at the ceramic/PANI interface which results in the increase of interfacial polarization [21]. The intense conductivity of the composites may be due to the addition of conducting polymers [22]. Furthermore, polarons in PANI chains enhance the conductivity of the composites. Decrease in resistivity (R) of the composites with increasing frequency was observed as shown in Figure 6. The decrease may be due to the upgrading of charge carriers' migration in the materials which further enhances their conductivity. Decrease in resistivity also represents the semi-conductivity of the materials. C2P composite displays highest resistivity ( $R = 2.07 \times 10^2 \Omega \cdot \text{cm}$ ) relative to other two composites, which inversely decreases its conductivity  $(0.007 \Omega \cdot \text{cm})^{-1}$ . Another reason for its low conductivity is its lowest dielectric loss value. Due to high conductivity, these materials are preferred to be used as semi-conductors.

To know about the capacitance (C) performance of the composite materials, pellets of size 5mm–10mm of the prepared materials were made and were adjusted in between metal electrodes. The two electrodes

were connected to a voltage source as shown in the schematic diagram in Figure 7. The particles of the composites get polarized in the field and line up themselves in a way that sets up another field inside, which is opposite to the field of the capacitor plates. Capacitance of all composites is decreasing with increasing current frequency from MHz to GHz. C1P and C3P have  $6.49 \times 10^{-11}$  and  $5.13 \times 10^{-11}$  F capacitance respectively while C2P has a capacitance of 2.95 pF. It is ascribed to the wide space charge regions in C2P as well as the absence of so many metal ions in C2 as confirmed by EDX analysis (Table 1). At about 2 GHz, capacitance exhibits resonance phenomena while at 2.5 GHz, an immediate drop occurs which may be due to the partially blocked charge carriers in the composites near the electrodes surface [22].

### 3.6. Mechanical properties

Prepared samples were tested for rheological characterization at different temperatures. Mechanical rigidity and stability of a material is represented by the storage modulus  $G'$  @ angular frequency. The  $G'$  value rises with increasing angular frequency as shown in Figure 8a, b and c. The composites exhibit  $G'$  in the range of  $5.10 \times 10^3$  Pa to  $5.37 \times 10^3$  Pa, which represent the elastic nature of the materials. The materials are stiff as shown by the high  $G'$  value [23]. The interactions were further confirmed by the SEM analysis.  $G'$  value shows the energy storage capability of these materials. Hence these materials are considered to be best suited where damping or piezoelectric properties are required.

Complex viscosity  $[\eta^*]$  analysis was done to find the flow behavior of the composites in support of mechanical properties at different temperatures. The rheograms (Figure 9a, b, c) show that complex viscosity for C1P, C2P and C3P first declines up to  $80 \text{ s}^{-1}$  after which a sudden increase is observed which specifies the relaxation of polymer chains in the composites and hence the free mobility of ceramic particles in the PANI matrix is restricted further. This also implies that the materials exhibit shear thinning behavior at low frequency and then an upshot in the viscosity with increasing frequency shows shear thickening [20]. The interactions among the C1P, C2P and C3P ions/dipoles are weak but with increasing frequency, the dipoles attract each other through physical and chemical bonds and hence the materials adopt a viscous and dense structure.  $[\eta^*]$  value is in the range from 10.2 to 10.8 Pa.s. It is concerned with molecular vibrations and dipole rotation in the materials that results in increased dielectric losses and hence conductivity in the composites [24, 25]. Table 4 describes a summary of various dielectric and mechanical properties of the prepared materials in this study.

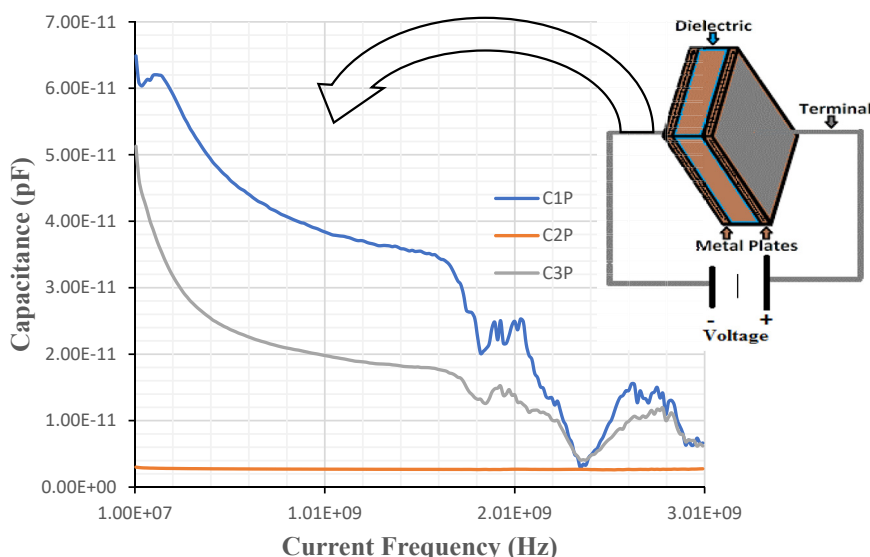
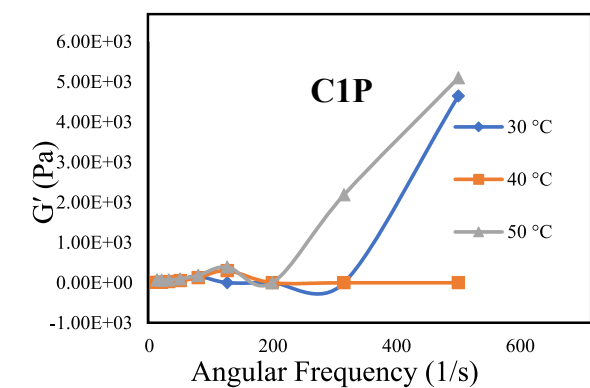
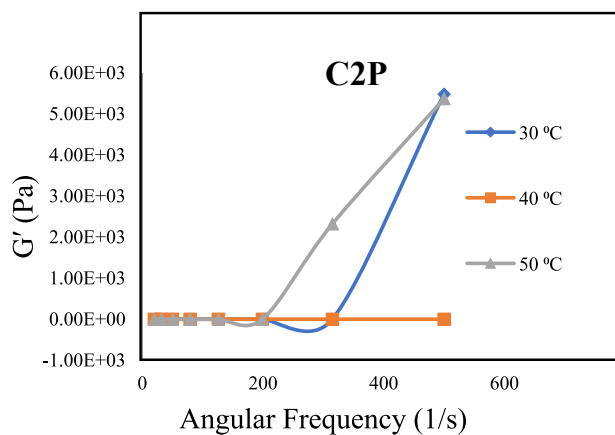


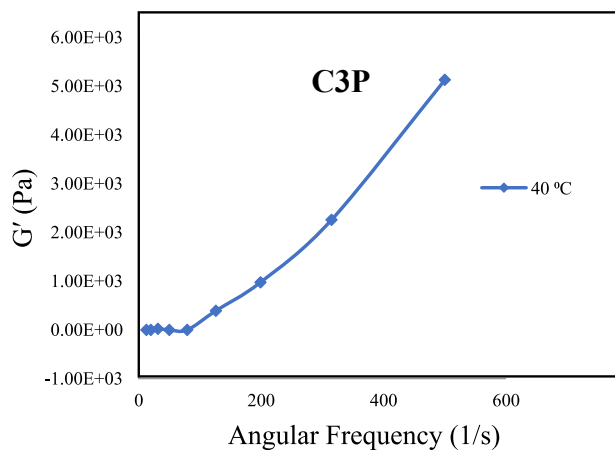
Figure 7. Composite based capacitors and their capacitance.



(a)



(b)

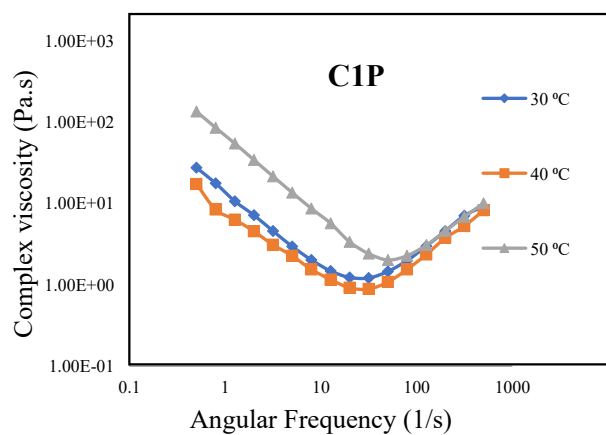


(c)

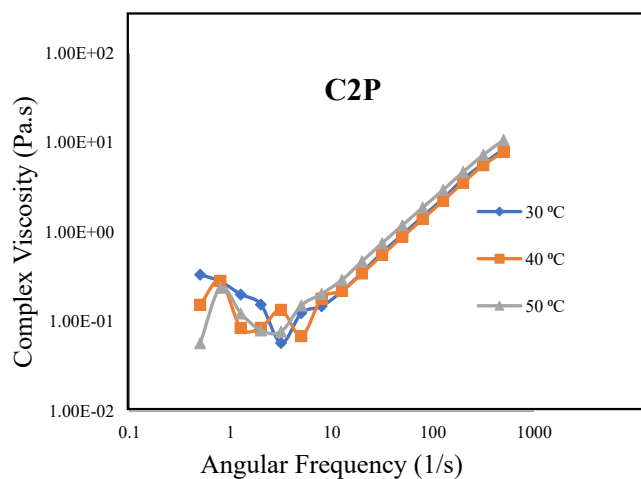
Figure 8. Mechanical storage modulus at different temperature: (a) C1P, (b) C2P and (c) C3P.

4. Summary

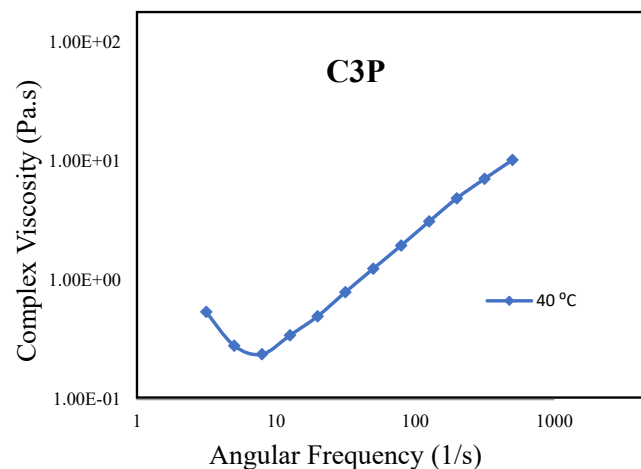
Ceramic wastes were collected from the premises of industrial area Hayatabad, Peshawar, KP, Pakistan. We analyzed waste ceramics based PANI composites which are novel materials in the sense that they have low density, high thermal stability, good dielectric properties and high mechanical strength. The dispersion of waste ceramics particles in polymer matrix and their composite formation were confirmed by FT-IR analysis and SEM. The composites were used in capacitors where they showed capacitance in pF. The materials showed high *ac* conductivity ( $3.67 \Omega^{-1} \text{cm}^{-1}$ ) and mechanical strength ( $G' = 5.37 \times 10^3 \text{ Pa}$ ). These



(a)



(b)



(c)

Figure 9. Complex viscosity as a function of angular frequency: (a) C1P, (b) C2P and (c) C3P.

characteristics of the novel composites clearly demonstrate their possible application in semi-conductors, microwave devices, capacitors, thermal insulation and as piezoelectric materials. Reuse of ceramic wastes for

**Table 4.** Summary of dielectric and mechanical properties of the composites.

Samples	$\epsilon'$	$\epsilon''$	$\tan\delta$	$C_p$ (F)	$\sigma$ ( $\Omega\text{.cm}$ ) <sup>-1</sup>	R ( $\Omega\text{.cm}$ )	G' (Pa)	$[\eta^*]$ (Pa.s)
C1P	61.1	253	6.14	$6.49 \times 10^{-11}$	3.80	3.52	$5.10 \times 10^3$	10
C2P	3.60	0.23	0.17	$2.91 \times 10^{-12}$	0.01	-do-	$5.37 \times 10^3$	10
C3P	93.6	922	129	$5.13 \times 10^{-11}$	3.29	-do-	$5.13 \times 10^3$	10

good purposes make them better than worse. This may be an alternative disposal and good solid waste management while reducing their burden upon the environment in the mentioned area.

### Declarations

#### Author contribution statement

M. Sohail: Conceived and designed the experiments.  
 Sana Ullah Khan, M. Saleem Khan: Conceived and designed the experiments; Wrote the paper.  
 Ihsan Ullah, M. Omer, Adnan: Analyzed and interpreted the data.  
 Noor Saeed, Sabiha Sultana: Performed the experiments.  
 Adnan Shahzad, Mian Gul Sayyed: Contributed reagents, materials, analysis tools or data.

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#### Data availability statement

Data will be made available on request.

#### Declaration of interests statement

The authors declare no conflict of interest.

#### Additional information

No additional information is available for this paper.

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