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**Swagatika Mishra[1], Punyapriya Mishra[1*], Punyatoya Mishra[2], Deepak Kumar Mohapatra[1], Dinesh Kumar Mishra[1] Krushna Prasad Shadangi[1]**

*[1] Veer Surendra Sai University of Technology, Burla, Odisha, India 768018*
*[2] Parala Maharaja Engineering College, Berhampur, Odisha, India 761003*
**Corresponding author: E-mail: ppmishra_me@vssut.ac.in*

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# STUDY OF MECHANICAL AND MORPHOLOGICAL PROPERTIES OF CCTO-BT/EPOXY COMPOSITE

**The demand for environment-friendly ceramic reinforced polymer matrix composite (CRPMC) fabrication leads to the development of lead-free CRPMC. Calcium copper titanate (CCTO) and barium titanate (BT) are two of the most widely used lead-free ceramics for embedded capacitor applications. In the present study, the mechanical and morphological properties of both single and hybrid ceramic (CCTO and BT) filled epoxy composites were evaluated and compared with the unfilled pure epoxy resin. Hand lay-up followed by the compression molding technique were used to synthesize the CRPMC samples. Among the single filler CRPMCs, the BT/epoxy composite exhibited better mechanical properties and density values than the CCTO/epoxy composite. The 60:40 ratio hybrid CCTO-BT/epoxy composite possessed the highest mechanical properties and density values in contrast to the other composite specimens. The SEM micrographs of the fractured surfaces of the BT and CCTO CRPMC specimens were found to have a rougher and wavier appearance than the unfilled epoxy.**

**Keywords:** **CRPMC, lead-free ceramic, epoxy, SEM, mechanical properties**

## INTRODUCTION

In the modern world, the rapid growth in the applications of electronics drives researchers to investigate further in this field. In electronics applications, there is a great demand for small, and complex-shaped products than for structural applications [1-3]. The growing interest in enhancing the electrical performance of electronics systems leads to an increase in the ratio of passive to active components. For example, the ratio of passive to active components in cellular phones usually is over 20 [4]. The discretized surface mounting of passive components over the substrate interface faces several challenges like large substrate area consumption, reliability issues with multiple solder joints, and lower electrical performance owing to an extended interconnection length. Embedded passive technology has been increasingly investigated to develop passive components through multi-layered substrates to address the challenges. In this context, embedded capacitors have a wide number of applications, including by-pass, tuning, signal decoupling, and filtering. The major requirements for embedded capacitors include easy processability, low processing cost, low capacitance tolerance, and excellent dielectric performance. The ceramic reinforced polymer matrix composite (CRPMC) is a potential candidate for embedded capacitor applications that collectively process a flexible polymer and a ceramic with improved dielectric properties [5].

CRPMC processing has a comparatively lower cost, requires a lower processing temperature, and has excellent compatibility with multi-layered organic substrates [5-7]. Improvement in the desired properties of CRPMCs depends on the filler loading, filler material, interfacial bonding between the matrix and reinforcement phase, method of dispersion, the filler distribution and filler size, as well as its shape [3, 8-15]. The epoxy polymer resin is a preferable polymer resin over other polymers for CRPMC synthesis because of its excellent ability to form a strong covalent cross-link. The most commonly used ceramic materials are lead based ferroelectric ceramics such as PZT (lead zirconate titanate) and its derivatives. In addition, the use of epoxy is also responsible for the synthesis of void-free composites [16, 17]. Concerning environmental issues, lead-free ceramics such as barium titanate (BT) and calcium copper titanate (CCTO) are gaining more interest than lead-based ceramics as reinforcement material. BT and CCTO are well-known reinforcement materials for CRPMC development for embedded capacitor applications due to their excellent dielectric performance. The literature on CRPMC development confirms that the increase in the ceramic filler content is responsible for the substantial advancement in the dielectric properties. On the other hand, it also deteriorates the mechanical properties. Hence, for applicability purposes, evaluation of the mechanical properties of developed CRPMCs is highly essential to determine the service life and usefulness. Sathish et al. [11] investigated the effect of SiC and $Al_2O_3$ additions on the morphology and mechanical properties of flax reinforced epoxy composites. Varied SiC and $Al_2O_3$ reinforcement between 2-10 wt.% revealed that the composite with 8 wt.% SiC and 2 wt.% $Al_2O_3$ exhibited the highest mechanical properties, including tensile strength, interlaminar shear strength, flexural strength, and impact strength. Beyond 8 wt.% SiC, the SiC particles tended to agglomerate, which led to poor interfacial bonding between the fillers and fibers with the epoxy matrix and to reduced mechanical properties. Luangchuang et al. [18] reported that an excessive increase in CCTO ceramic filler loading in acrylonitrile-butadiene rubber (NBR) leads to particle agglomeration. The agglomeration of the CCTO particles results in poor filler-rubber interactions, causing degradation in the tensile properties. Poor dispersibility and compatibility of the ceramic filler particles in the polymer matrix are also responsible for the partial mechanical breakdown of CRPMCs. Earlier research on ceramic reinforcement in a hybrid form can eliminate intrinsic flaws without sacrificing the advantages of CRPMCs. In the hybrid reinforcement of BT and CCTO, the difference in the span factor and size between these two ceramic particles significantly enhances the dispersibility of the fillers in the polymer matrix to improve the mechanical properties of CRPMCs [9, 10, 19]. The smaller-sized BT particles filled up the interstitial voids among the larger-sized CCTO particles to produce a packed structure.

Few publications are available on the evaluation of the mechanical properties of BT and CCTO reinforced epoxy polymer matrix composite. Saidina et al. [10] observed a BT reinforced epoxy composite (BEC) showing comparatively better tensile properties than a CCTO reinforced epoxy composite (CEC). This improvement results from the smaller span factor of BT, which leads to homogeneous distribution of BT across the epoxy matrix with a smaller void content in BEC. The present study aims to analyze different mechanical properties, including tensile strength, compressive strength, flexural strength, and hardness, along with the density of the unfilled epoxy in addition to both single and hybrid filler (BT and CCTO) loaded epoxy matrix--based composites. Furthermore, SEM micrographs of the tensile-tested fractured surface of the specimens were analyzed precisely to evaluate the morphological properties.

## MATERIALS AND METHODS

### Materials

The CCTO and BT ceramic powder samples were synthesized by the conventional solid-state reaction route. The starting materials for CCTO and BT ceramic preparation with 98% purity were supplied by LOBA Chemie pvt Ltd. For CCTO ceramic preparation, $CaCO_3$, CuO, and $TiO_2$ ceramic powders were used as the starting materials, whereas $BaCO_3$ and $TiO_2$ ceramic powders were employed as the starting materials for the BT ceramic preparation. Bisphenol A diglycidyl ether (LY 556) was used as the epoxy resin procured from Sambalpur, Odisha, India. HY 951 (triethylenetetramine) was utilized as the hardener, supplied by Ciba-Geigy of India Ltd.

### BT-CCTO/epoxy composite preparation

The moisture content from the precursors was extracted by heating them in a digital oven to 110°C for 1 h. The precursors were mixed in a high energy planetary ball mill to obtain a homogeneous mixture in the presence of ethanol as grinding media with 5 mm diameter WC balls for 4-5 h. Subsequently, the mixtures were dried, followed by calcination. For CCTO ceramic preparation, the calcination temperature was taken as 1000°C for 12 h, and for BT ceramic preparation, it was at 1200°C for 6 h. Eventually, the prepared BT and CCTO ceramics were ground to powder form with the help of a mortar pestle for 3 h. The sonication of the DGEBA type (LY 556) epoxy resin was done at 60-70°C to reduce its viscosity. Afterwards, the ceramic powders (BT and CCTO) were added in different weight ratios to the sonicated epoxy polymer resin, where triethylenetetramine (HY 951) was employed as the hardener. The ratio of ceramic reinforcement to polymer matrix was taken as 20:80 for all the different weight ratios of single and hybrid fillers. The samples were fabricated at five different weight ratios of filler materials (CCTO and BT ceramic powders): 100:0, 0:100, 50:50, 60:40, 40:60. The liquid mixture was stirred for 10-15 minutes for uniform mixing at room temperature, then was poured into a silicone spray coated stainless steel mold with the dimensions 180 mm × 180 mm × 3 mm and was kept at room temperature for 24 h. Afterwards, it was post-cured at 120°C in a compression molding machine for 2 h under a pressure of ~70 kg/cm$^2$. The samples were cooled down to room temperature and subsequently cut into different dimensions as per the requirements to characterize them.

### Characterization

The characterization of the pure epoxy as well as all the single and hybrid ceramic filled epoxy composites were carried out to evaluate and compare the mechanical and morphological properties. The tensile test and three-point bending test were performed by means of an INSTRON 3382 UTM. The tensile test was conducted to evaluate the tensile strength, elongation at break, and tensile modulus values of the composite specimens. Furthermore, SEM (SU3500, Hitachi High-Technologies Corporation) observations of the fractured specimens after the tensile test were performed to characterize the morphological properties. The three-point bending test was carried out to determine the flexural strength and modulus of the composite specimens. Vickers hardness measurements of the composite samples were obtained using a universal hardness tester (251 VRSA, AFFRI). A density meter was used to determine the density of the composite specimens using Archimedes' principle. Compression tests of the composite specimens were performed using a UTM UTE 20 HGFL to determine the compressive strength.

## RESULTS AND DISCUSSION

The tensile test of the prepared composite specimens was performed to determine the tensile strength, modulus, and elongation at break values. In addition, SEM investigations of the fractured surfaces of the tested specimens were performed to analyze the topographical features and thereby unveil the root cause of the fracture. Figure 1 shows the different tensile plots of the pure epoxy resin, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens. The variation in the tensile strength and modulus of the CRPMCs with the variation in the weight fractions of BT and CCTO in the epoxy matrix can be observed in Figure 1a. In contrast to the unfilled epoxy composite, the tensile strength and modulus of the other composite specimens show an increasing trend owing to the incorporation of the BT and CCTO ceramic particles into the epoxy matrix phase. Moreover, the hybrid reinforcement of both BT and CCTO subsequently enhances the tensile properties of the CCTO-BT/epoxy composites.

The tensile strength and modulus of the pure epoxy resin were found to be 8.77 MPa, and 1650.9 MPa, respectively, which are the lowest among all the other single and hybrid filler reinforced epoxy composites. Furthermore, among the single CRPMCs, the CCTO/epoxy composite exhibits lower tensile strength and modulus of 20.17 MPa, and 1788.48 MPa, respectively, compared to the BT/epoxy composite. This is mainly because of the agglomeration of the filler material, lacking in interfacial adhesion between the filler and the matrix phase, and a significant rise in the stress concentration [20]. In addition, the smaller span factor and compatibility of the BT ceramic with the epoxy matrix than CCTO leads to higher tensile properties of the BT/epoxy composite [10]. The BT/epoxy composite is comparatively stiffer than the CCTO/epoxy composite since stiffness is proportional to the tensile modulus. Hence, it can be confirmed that the single filler reinforcement of BT in the epoxy matrix makes the composite stiffer. On the other hand, the hybrid ceramic filler reinforced 60:40 ratio based CCTO-BT/epoxy composite was found to have the highest tensile strength and modulus of 36.54 MPa, and 2100.29 MPa, respectively, in contrast to the unfilled epoxy resin, single filler reinforced epoxy and the other hybrid filler reinforced epoxy composite specimens. In the composite with the hybrid filler reinforcement, the smaller BT particles occupy the interstitial voids present in between the larger CCTO particles [10], thus leading to significant improvement in the tensile properties.

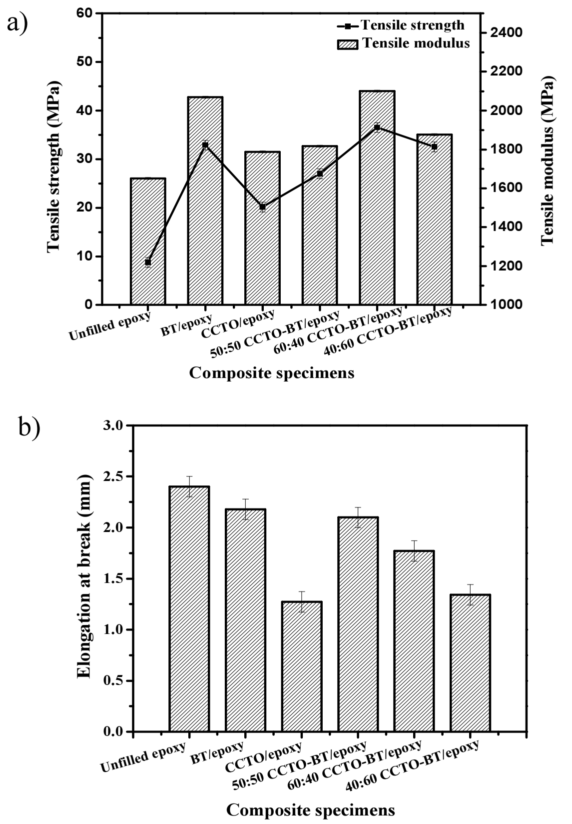


Fig. 1. Comparative tensile test plots of unfilled epoxy and CCTO-BT/epoxy composites: a) tensile strength and tensile modulus, b) elongation at break

The elongation at break value for the unfilled epoxy resin was found to be 2.4 mm, the highest among all the other single and hybrid filled specimens (Fig. 1b). It may be a consequence of the high epoxy/filler mixture viscosity in the single filler (CCTO/epoxy and BT/epoxy) and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens that led to an increase in contact between the filler particles. What is more, an increase in the particle-particle contact between the filler particles leads to an rise in the void content and filler agglomeration, which affect the elongation at break values [10]. Moreover, the reduction in interstitial void spaces between the CCTO particles by means of BT particles in the hybrid filler filled epoxy composites might be responsible for the intermediate elongation at break values of the CCTO-BT/epoxy composite specimens.

Figure 2 presents the SEM micrographs of the fracture surfaces of the pure epoxy resin, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO--BT/epoxy) reinforced epoxy composites. The SEM micrograph of the pure epoxy resin reveals that it has a very smooth surface appearance, which indicates it to be a brittle type of fracture [10, 21]. It is also inferred that the unfilled epoxy has very low resistance to crack propagation and can easily break by applying load [10]. On the other hand, the fracture surface of the CRPMC specimens have a rougher and wavier appearance than the unfilled epoxy, which infers that the energy required for crack propagation in the case of ceramic reinforced epoxy composites is more than that of the pure unfilled epoxy composite [10, 22, 23]. Uniform distribution of the ceramic particles in the epoxy matrix can be observed in the figure. With the increase in the volume fraction of ceramics, the connectivity of the ceramics grows, which confirms 0-3 connectivity. In 0-3 connectivity-based composite, the 0-dimensional filler particles are reinforced in a 3-dimensional continuous matrix phase.

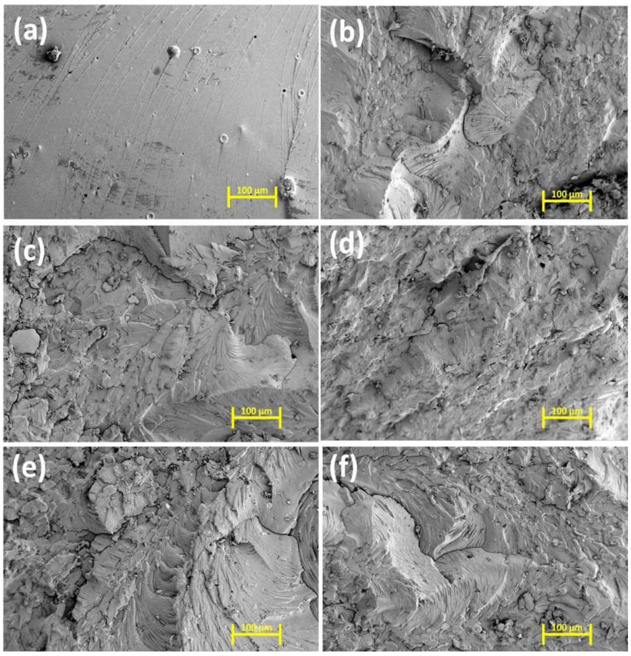


Fig. 2. SEM micrographs of fracture surfaces of: a) pure epoxy, b) BT/epoxy, c) CCTO/epoxy, d) 50:50, e) 60:40, f) 40:60 CCTO-BT/epoxy composites

Figure 3 shows the variation in the flexural strength and modulus of the pure epoxy, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens. The variation in the flexural properties of the composite specimens shows a very similar kind of trend to the tensile properties. In contrast to the unfilled pure epoxy resin, there a significant increment in the flexural strength and modulus for both the single filler (CCTO/epoxy and BT/epoxy) and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens (Fig. 3). The flexural strength and modulus values of the unfilled epoxy resin were obtained as 7.49 and 60.58 MPa, respectively, as the lowest in comparison to the other single and hybrid filler reinforced epoxy composites. Like the tensile properties, the BT/epoxy composite was also found to have higher flexural strength and modulus of 26.28 and 3383.24 MPa, respectively, in comparison to the CCTO/epoxy composite as a result of the smaller span factor and the compatibility of the BT ceramic with the epoxy matrix phase. The hybrid filler reinforced 60:40 ratio CCTO-BT/epoxy composite possesses the highest flexural strength and modulus of 31.12 and 3587.29 MPa, respectively, in contrast to the unfilled epoxy resin, single filler reinforced epoxy, and other hybrid filler reinforced epoxy composite specimens. It is mainly because of the enhancement of interfacial adhesion or bonding between the reinforcement (BT and CCTO) particles with the epoxy matrix. Moreover, better particle distribution and filler packing due to the reduction in the interstitial void spaces among the CCTO ceramic particles by the smaller BT particles are also responsible for significant improvement in the flexural properties of the hybrid CCTO-BT/epoxy composites [24].

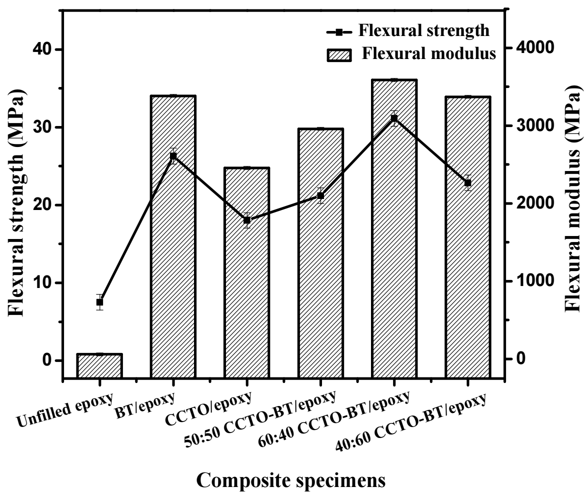


Fig. 3. Comparative plots of flexural strength and modulus of epoxy and CCTO-BT/epoxy composites

Figure 4 displays the variation in the compressive strength of the pure epoxy resin, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens. The comparative compressive strength plots reveal that both the single and hybrid filler reinforced epoxy composites have higher compressive strength than the unfilled epoxy resin (Fig. 4) as in the tensile and flexural properties plots (Figs. 1 and 3). The interfacial bonding and amount of voids in the composite specimens influenced the compressive strength to a great extent. The 60:40 ratio CCTO-BT/epoxy composite has the highest compressive strength of 160.17 MPa among the unfilled epoxy resin, all the single and hybrid filler reinforced epoxy composites. The enhancement of the interfacial bonding between the two different sized ceramic particles and the ceramic-polymer interface is mainly responsible for the significant growth in compressive strength.

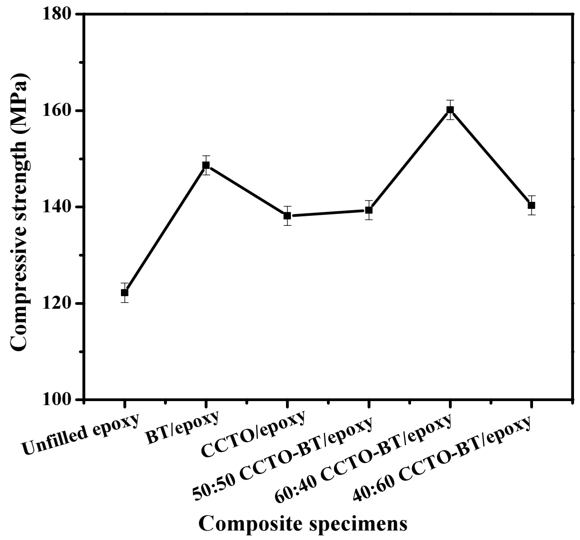


Fig. 4. Comparative compressive strength plots of unfilled epoxy and CCTO-BT/epoxy composites

Figure 5 presents the Vickers hardness (HV) values of the pure epoxy, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens. The hardness values for both the single filler (CCTO/epoxy and BT/epoxy) and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens show a rising trend in contrast to the unfilled pure epoxy (Fig. 5). It is because the incorporation of filler particles that the composite is more rigid. As a result, under the equivalent rate of indentation loading, the filler particles collide with each other more quickly to effectively circulate the accumulative stress. Henceforth, the composite withstands a higher stress level before undergoing plastic deformation [24, 25]. In Figure 5, it can be observed that the hybrid filler reinforced 60:40 ratio CCTO-BT/epoxy composite has the highest microhardness value of 34.9 HV, in contrast to the unfilled epoxy resin, single filler reinforced epoxy, and the other hybrid filler reinforced epoxy composite specimens. This may be due to the presence of a more packed structure of filler particles in the epoxy matrix, which strengthens the composite structure [24]. Furthermore, the differing particle sizes of CCTO and BT collide with each other in a more rapid manner under the indentation load to cause effective distribution of the accumulative load before plastic deformation, which helps to resist a higher magnitude of stress, and hence a higher hardness value [24, 25].

Figure 6 shows the variation in the experimental density of the pure epoxy resin, single filler (CCTO/epoxy and BT/epoxy), and hybrid filler (CCTO-BT/epoxy) reinforced epoxy composite specimens. It is apparent that the introduction of the ceramic filler increases the density of the epoxy composites. In the case of the single filler reinforcement, the BT/epoxy composite has higher density compared to the CCTO/epoxy composite owing to the low span factor of $BaTiO_3$, which indicates that its small particle size is responsible for the dense packing. Moreover, the 60:40 ratio CCTO-BT/epoxy composite exhibited the highest density of 1.42 gm/cm$^3$ (Fig. 6) in comparison to the unfilled epoxy resin, single filler reinforced epoxy, and other hybrid filler reinforced epoxy composite specimens. This high density value of the hybrid 60:40 ratio CCTO-BT epoxy composite results from the denser packing of the filler particles in the epoxy matrix. Therefore, the different sizes of the BT and CCTO particles effectively produced a densely packed structure.

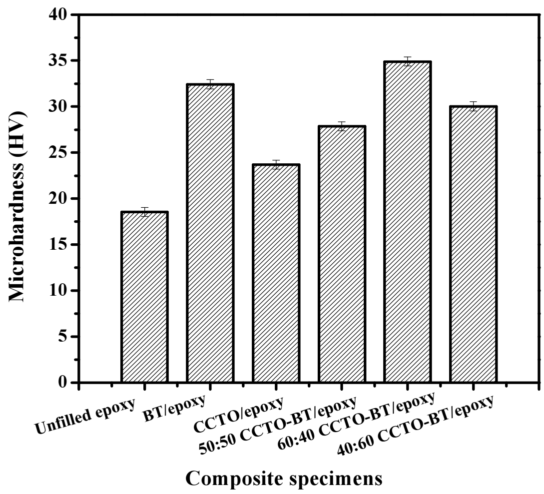


Fig. 5. Comparative plots of Vickers hardness (HV) of unfilled epoxy and CCTO-BT/epoxy composites

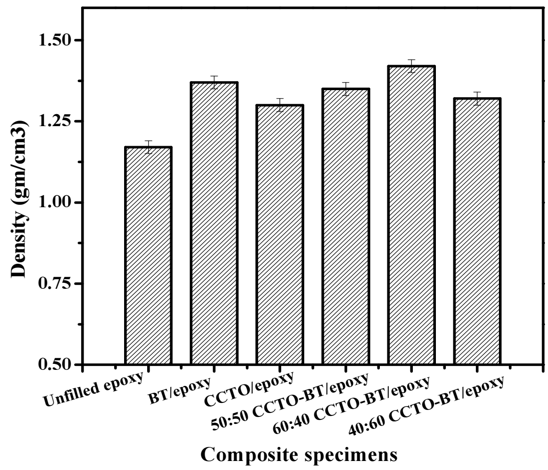


Fig. 6. Comparative plots of density of unfilled epoxy and CCTO--BT/epoxy composites

## CONCLUSION

The following conclusions were drawn from the present study:

- Both the single and hybrid filler reinforced epoxy composites exhibited significant improvement in the density and mechanical properties including the tensile properties, flexural properties, hardness, and compressive strength.
- The SEM micrographs of the fracture surfaces of the BT and CCTO CRPMCs revealed a rougher and wavier appearance than the unfilled epoxy.

 

– Between the single reinforcement CRPMCs, the BT/epoxy composite has higher mechanical properties and density than the CCTO/epoxy composite.

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