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DC electrical conduction mechanism in polypropylene–Bijoypur white clay (PP-BWC) composites

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Abstract : Polypropylene-Bijoypur white clay (PP-BWC) composites were prepared at different compositions of PP and BWC of Bangladesh by extrusion technique. Structural change was investigated using X-ray diffractometry (XRD). The dc electrical conduction mechanism of different PP-BWC composites was studied by current-voltage (I–V) characteristic measurements. It is observed that Poole-Frenkel type of mechanism is operative in these materials. The dc conductivity measurement of the different samples was performed in the temperature range 298 K to 400 K. The temperature dependence of electrical conductivity indicates that the conduction is due to thermally activated hopping of charge carriers.

Keywords : Polypropylene-Bijoypur white clay composites, X-ray diffractometry, dc electrical properties

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1. Introduction

Polymer composites have attracted much attention of the researchers in recent years because of their application in a variety of areas such as electrostatic charge dissipation, electromagnetic interference/radio frequency (EMI/RF) shielding, electrodes for polymer-based batteries and intermediate layers of high-voltage cable [1–5]. Many reports have appeared both on the theory of electrical conduction [6,7] and experimental finding in a number of such composites [8,9]. Radhakrishnan and Saini [10] carried out an investigation on dc and ac conduction of polyester elastomer filled with various metallic and ferrite fillers as a function of temperature, frequency and filler concentration. The results have been explained on the basis of Schottky barrier formation and the depletion regions formed near the polymer-metal interface. There are also reports on the electrical properties of ceramic/polymer composites [11], mixed rubber composites [12], polyethylene-carbon-fiber composites [13], carbon black-filled high density polyethylene (HDPE) composites [14], polyaniline/polypropylene (PANI/PP) composites [15]. The dielectric loss of the ceramic/polymer composites is observed to dominate by those of the polymer, whereas the ceramic phase may have a significant contribution on the steady-state electrical conduction and low-frequency dielectric loss at high temperature [11]. The activation energy decrease observed in mixed rubber composites may be associated with gradual decrease in the temperature coefficient of conductivity [12].

Shan Ren and Hon [16] prepared composite from a newspaper fiber-reinforced polypropylene (NPF/PP) and interfacial modification on the mechanical properties were investigated. Natural fibers, such as recycled corrugated board, chemical wood pulp, thermomechanical pulp, and cotton pulp were also found to have reinforcing properties for polypropylene. The mechanical properties have been improved by blending of carbon fiber with the NPF/PP composites. Ahmad *et al* [17] have investigated the electrical

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properties of polypropylene-talc (PP-talc) composites. It was found that electromagnetic waves interact with the material *via* the impurities, inclusions and voids existing in the bulk composites. The impedance, return loss and insertion loss shows relatively low frequency dependence. These results suggest that this composite material may find some microwave applications.

Bijoypur white clay (BWC) is being used in ceramics, insulators and sanitary industries of Bangladesh. Chowdhury and Bhuiyan carried out investigations on the structural [18] and dc electrical [19] properties of BWC on heat treatment at high temperature. But so far, no study of the electrical properties of PP-BWC composites has been appeared in the literature.

This work is an attempt to elucidate the dc electrical properties of PP-BWC composites from the current-voltage (I-V) characteristics and temperature dependence of dc electrical conductivity. The XRD analysis of the composites is also included.

2. Experimental details

2.1. *Sample preparation :*

Composites used in this work were prepared from PP (NOVOLEN 1100L 2510025, BASF, Germany, density = 900 kg/m^3, melting temperature = 338 K, glass transition temperature = 270 K, refractive index $\mu = 1.3$, molecular weight M_w = 184700 g/mole) and uncompressed hard and solid BWC collected from mine at Bijoypur, Netrakona, Bangladesh. Collected BWC lumps were manually crushed into powder using mortal and pestle and then sieved through a 200 mesh sieve. At first, five batches with different compositions (wt%) of PP and BWC powder were prepared according to the formula 10PP : xBWC where x = 1, 2, 3, 4, 5. The samples are represented as

Sample designation	A	B			C_3	C_4	C_5
Composition (wt%)	PP	BWC	10 : 1	10 : 2	10 : 3	10 : 4	10 5

The different mixtures were mixed during passing through an extrusion machine. The molten material was collected through the die in the form of rod. The rod shape composite samples were cut into pellets. The samples were then placed inside a die punch of internal diameter 0.019 m. The pellet shape samples were pressed using a hydraulic pressing unit at a pressure of 34 MPa at 398 K for five minutes. After removing the sample from the die punch, its upper and lower faces were polished and then thin coating of conducting silver paste was applied on both sides which act as electrode for electrical measurements.

2.2. *Measurement techniques :*

2.2.1. *X-ray diffraction*

An X-ray diffractometer (Model JDX-8P JEOL Ltd., Tokyo, Japan) was used to record the XRD patterns of all the samples. The target is Cuk$_\alpha$ and the wavelength of X-ray radiation is 1.5418 Å. The XRD patterns of the samples mentioned in Section 2.1 were recorded as a function of 2θ from 3 to 73°.

2.2.2. *DC electrical measurement*

The dc current was measured using a digital Keithley Electrometer (model 614, Keithley Instrument Inc., Cleveland, Ohio, USA). A pressure contact specimen holder housed in a steel chamber, with an in-built heater was employed for the measurement. A transistorized power supply, model 6181C of Hewlett Packard was used for voltage supply across the samples. The dc conductivity was calculated from the measured dc current through the sample and effective sample dimensions. Thickness of the sample is 4.25×10^{-4} m and diameter is 6.35×10^{-3} m. The I-V characteristic measurement was performed from 10–160 V at room temperature. The temperature dependence of dc current was measured from 298 K to 420 K with a constant dc applied voltage of 100 V. The temperature was recorded by a Cr–Al thermocouple by measuring the emf with a microvoltmeter (model 197A of Keithley Instrument Inc. Cleveland, Ohio, USA). All the measurement were performed in vacuum ($\approx 10^{-3}$ torr).

3. Results and discussion

3.1. *XRD analysis :*

X-ray diffraction patterns of the samples A, B, C_1, C_2, C_3, C_4 and C_5 are presented in Figure 1. These diffraction

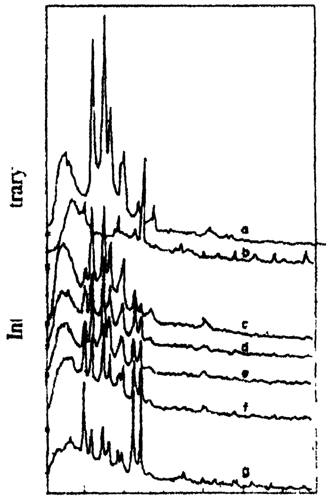

Figure 1. The XRD patterns of all the samples represented by (a) : A (PP), (b) : B (BWC), (c) : C_1, (d) : C_2, (e) : C_3, (f) : C_4 and (g) : C_5

patterns are represented respectively by diffractograms *a*, *b*, *c*, *d*, *e*, *f* and *g*. The details of the X-ray scans (2θ and d_{hkl}) are depicted in Table 1.

Table 1. 2θ and d_{hkl} values of samples A (PP) and B (BWC).

PP		BWC	
2θ (degree)	d_{hkl} (Å)	2θ (degree)	d_{hkl} (Å)
7.2	12 277	9.2	9 612
14 0	6 325	12.6	7 030
17 0	5.215	20 0	4.440
18 6	4.770	21.0	4 230
21 9	4 058	25.0	3.560
25 6	3 479	26 8	3.320
29 4	3 038	36 6	2 455
33.8	2.652	38.0	2 370
43.4	2 085	39.4	2 286
47 8	1 903	42.4	2.132
48.8	1.866	45.8	1 981

In the XRD pattern of sample A (PP), it is observed that the intense diffraction maxima appear at the 2θ values of 14°, 17°, 18.6°, 21.9°, 25.6°, 29.4°, *etc*. These values are matched very closely with the values reported by several authors [20,21] indicating that the PP used in this experiment is isotactic. The X-ray scans of PP indicate monoclinic α form crystallites with some peaks absent. The absence of some peaks could be due to orientation of the crystalline phase [22].

The XRD pattern of the sample B (BWC) shows the main position of diffraction peaks at the 2θ values of 12.6°, 20°, 21°, 25°, 26.8°, 36.6°, 38°, 39.4°, 42.4°, 45.8°, 55°. The XRD analysis shows the presence of kaolinite (7.03, 3.56 and 1.45 Å), silicon oxide (4.44, 2.44 and 1.45 Å), iron oxide (1.67, 1.38 and 1.28 Å), potassium aluminium silicate hydroxide (4.93, 3.56, 2.53 and 2.37 Å), aluminium silicate (9.61 and 4.44 Å), *etc*. Very small amount of oxides of potassium, calcium, sodium, *etc* may also be present as reported by Biswas and Basak [23] and Chowdhury and Bhuiyan [18].

The XRD patterns of samples C_1 to C_5 containing different amount of BWC shown in Figure 1, exhibit most of the major characteristic reflections present in the XRD patterns of PP and BWC. It is observed that the intensity of the characteristic reflections (2θ = 12.6°, 20°, 21°, 25°, 26.8°, *etc*) of BWC is increasing and that of PP (2θ = 14°, 17°, 18.6°, 21.9°, 25.6°, 29.4°, *etc*) is decreasing gradually as the BWC proportion increases. The apparent doublet at 2θ = 21.9° in the XRD pattern of PP becomes a clear doublet and appears as two distinct intense reflections in case of composites with high BWC concentrations. These variations in the XRD reflections indicate a definite effect of BWC in the PP structure.

3.2. Current voltage (I–V) characteristics :

Figure 2 shows I–V characteristic curves for sample A, C_1, C_2, C_3, C_4 and C_5 measured at room temperature. It is observed that current increases non-linearly within the voltage range used. It is also seen that current increases as BWC concentration in PP increases. The non-linearity in I–V characteristics of PP and composites implies different conduction mechanisms in different voltage regions.

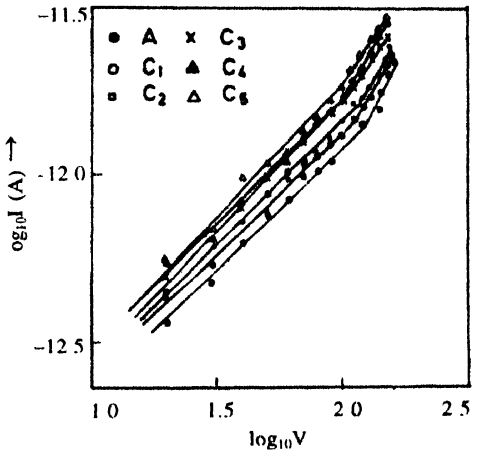

Figure 2. Variation of $\log_{10} I$ with $\log_{10} V$ (applied voltage in *V*) for different samples.

It is observed that the dependence of *I* on *V* can be fitted to a relation $I \propto V^n$, where *n* is the power factor. Various conduction mechanisms are characterized by different values of *n*. The case of $n = 1$ represents Ohmic conduction and is usually the result of the field-independent values of the carrier concentration and the mobility. The carriers may be ions, holes or electrons, but must be intrinsic to the material. If the value of *n* is more or less than unity, then the conduction mechanism is other than Ohmic. Here, there are two slopes : one at low voltage region 10–110 V, and the other at high voltage region 110–160 V, for all the samples. The *n* values are depicted in the Table 2. It is seen that the *n* values are close to unity in the lower voltage region and

Table 2. *n* values for different voltage regions for the samples A and C_1 to C_5.

Samples	Region 10–110 V	Region 110–160 V
A	0.75	1.66
C_1	0.75	1.45
C_2	0.75	1.40
C_3	0.75	1.25
C_4	0.80	1.20
C_5	0.80	1.10

that are much above unity at higher voltage region. The non-linearity of $\log_{10} I$ *versus* $\log_{10} V$ curves can be due to the space charge build-up giving rise to the non-uniformity of the field distribution between the electrodes and/or may also be related to the generation of carriers in the bulk of the materials. The above result suggests that the conduction mechanism in these materials may be space charge limited conduction (SCLC), Schottky or Poole-Frenkel type [24,25]. It is seen that as the BWC concentration increases, the difference in slope decreases. This means that the conduction mechanism tends to be independent of voltage at high BWC concentrations. It is interesting to note that composites with high BWC content show very close to Ohmic behaviour in the experimental voltage region.

The I-V data are plotted as $\log_{10} I$ *versus* $V^{1/2}$ in Figure 3 for further analysis. It is seen that the curves are linear in the higher voltage range. It is also observed that the $\log_{10} I$ *versus* $V^{1/2}$ curve becomes linear with the increase of BWC content throughout the voltage range. This suggests that the conduction is bulk limited. These results indicate the possibility of presence of Poole-Frenkel type of mechanism in these materials.

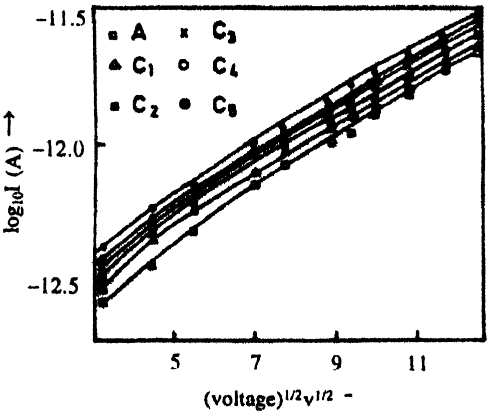

Figure 3. Variation of $\log_{10} I$ with $V^{1/2}$ (applied voltage in V) for different samples.

3.3. *Temperature dependence of dc electrical conductivity* (σ):

Figure 4 shows the plots of $\log_{10}\sigma$ *versus* $1/T$ curves for samples A, C_1, C_2, C_3, C_4 and C_5. It is seen that there are two temperature regions, namely, region I (298–360 K) and region II (above 360 K). The dc conductivity increases slowly in region I and that in region II increases rapidly with temperature.

The increase of σ with temperature may be due to the increased movement of the adventitious ions and/or electrons. The carriers experience a thermal velocity due to the increase in temperature in addition to the applied electric field which was kept constant at 100 V. So the resultant velocity experienced by the carriers is the sum of the drift velocity due to the applied field and the thermal velocity. In this study, the thermal velocity is changed due to the increase in temperature.

The activation energy (E_r) for the samples was determined from Figure 4. The values of activation energy associated with the two temperature regions of all the samples are depicted in Table 3. The activation energy of all the samples has higher value in high temperature region than those at low temperature region. In both low and high temperature regions, the activation energy decreases with increasing percentage of BWC. The existence of the shallow trap levels in the materials, is supported by the existence of different small values of activation energies at low and high temperatures. With the increase in temperature, carriers of respective trap levels take part in the conduction process.

The low activation energy as obtained from the temperature dependence of conductivity, indicates the existence of thermally activated hopping conduction in these

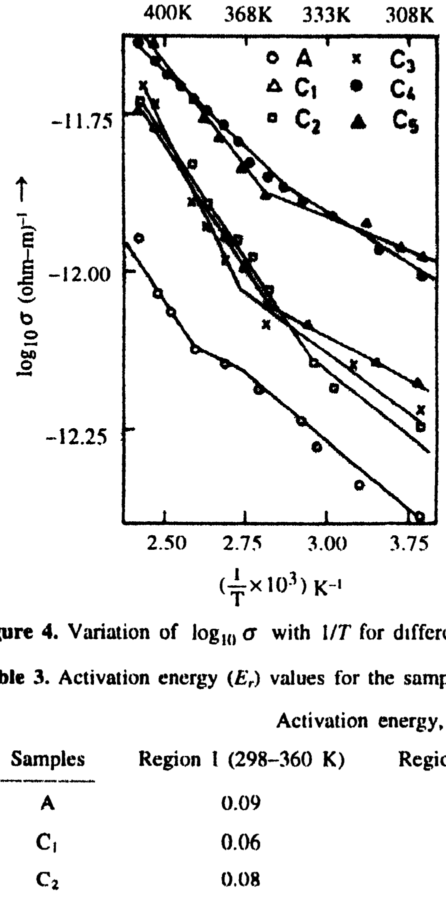

Figure 4. Variation of $\log_{10}\sigma$ with $1/T$ for different samples.

Table 3. Activation energy (E_r) values for the samples A and C_1 to C

Samples	Activation energy, E_r (eV) Region I (298–360 K)	Region II (above 360 K)
A	0.09	0.15
C_1	0.06	0.16
C_2	0.08	0.16
C_3	0.07	0.20
C_4	0.05	0.10
C_5	0.04	0.11

materials [26]. In this kind of conduction process, the carrier may be bound to the agglomerates itself. As a result, the carriers may not take part in the conduction throughout the bulk of the material.

4. Conclusions

The XRD investigation shows a definite change in the PP-BWC structure with BWC concentration. It may be inferred from the I–V characteristics that the Poole-Frenkel type of mechanism may be operative in these materials. The increase of dc electrical conductivity with temperature is due to increased movement of the adventitious ions and/or electrons within the material. The low activation energy indicates that the thermally activated hopping conduction is operative in these materials.

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