

# Negative Air Ion Releasing Properties of Tourmaline/Bamboo Charcoal Compounds Containing Ethylene Propylene Diene Terpolymer/Polypropylene Composites

Jen-Taut Yeh,<sup>1,2,3</sup> Han-Hsing Hsiung,<sup>3</sup> Wei Wei,<sup>2</sup> Ping Zhu,<sup>1</sup> Kan-Nan Chen,<sup>4</sup> Tao Jiang<sup>2</sup>

<sup>1</sup>Key Laboratory of Green Processing and Functional Textiles of New Textile Materials, Wuhan University of Science and Engineering, Ministry of Education, Wuhan, China

<sup>2</sup>Faculty of Material Science and Engineering, Hubei University, Wuhan, China

<sup>3</sup>Department and Graduate School of Polymer Engineering, National Taiwan University of Science and Technology, Taipei, Taiwan

<sup>4</sup>Department of Chemistry, Tamkang University, Tamsui, Taiwan

Received 10 May 2008; accepted 17 December 2008

DOI 10.1002/app.30003

Published online 2 April 2009 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** The average concentrations of negative air ions ( $C_{ion-}$ ) emitted from tourmaline (T), bamboo charcoal (B) particles, and tourmaline/bamboo charcoal (T/B) compounds containing polypropylene (PP) and ethylene propylene diene terpolymer/polypropylene (EPDM/PP) composite specimens under varying testing conditions were investigated in this study. The  $C_{ion-}$  values emitted from T or B filled PP and EPDM/PP composite specimens reached a maximum value as their T or B contents approached the 5 and 3 wt % optimum values, respectively. In contrast, the  $C_{ion-}$  values of T/B compounds filled PP and EPDM/PP composite specimens were significantly higher than their theoretical  $C_{ion-}$  values estimated using the "simple mixing rule," and reached a maximum value as the weight ratio of T to B reaches an optimum value. At this optimum T/B weight ratio, the  $C_{ion-}$  values of T/B compounds filled PP and EPDM/PP composite

specimens reached another maximum as their total compound loadings reached the optimum loading of 6 and 4 wt %, respectively. The  $C_{ion-}$  values of the PP/T/B and EPDM/PP/T/B specimens increased significantly as they were tested under dynamic mode or by increasing the testing temperatures. The T and/or T/B powders filled PP and EPDM/PP specimens exhibited significantly higher tensile strength ( $\sigma_f$ ) and elongation at break ( $\epsilon_f$ ) values than did the B filled PP and EPDM/PP specimens with the same filler loadings, respectively. Energy dispersive X-rays, particle size, and SEM morphology analysis of the filler particles present in the T, B, and T/B filled composite specimens were performed to understand these interesting negative air ion and tensile properties. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 113: 1097–1110, 2009

**Key words:** Composites; poly(propylene) (pp); resins

## INTRODUCTION

Negative ions present in air are generally recognized as being capable of exhibiting an invigorating effect on the living body by normalizing the autonomic or motorial nervous system. The person under the influence of negative ions is imparted with their biological functions such as sleep stimulation, ataraxy, activation of body cells, acceleration of metabolism, blood circulation, and fatigue recovery and so on.<sup>1,2</sup> For convenience, these negative ions present in air will be referred to as negative air ions. Presumably,

they can be present as varying types of negative air ions, such as  $O_2^-(H_2O)_n$ ,  $OH^-(H_2O)_n$ , or  $CO_4^-(H_2O)_2$ ,<sup>2,3</sup> when enough moisture contents are present in the ambient environment. Therefore, investigations of emitting negative air ions have drawn much attention for years.

Natural tourmaline ores can generate negative air ions spontaneously and permanently without generating excess amounts of radiation and ozone molecules.<sup>4</sup> It is worth noting that the efficiency of generating negative air ions of tourmaline crystals can be improved significantly by increasing their ambient temperatures or pressures, since they are generally recognized as pyroelectric and piezoelectric materials. It was also reported that the tourmaline mineral can emit far-infrared light, deodorize, purify water, and serve as effective antibacterial agent.<sup>5–8</sup> In view of the varying functional properties of tourmaline, investigations in preparing the new composite materials containing tourmaline capable of emitting negative air ions have drawn much attention for years. Many tourmaline-contained

Correspondence to: J.-T. Yeh (jyeh@tx.ntust.edu.tw).

Contract grant sponsor: National Science Council; contract grant numbers: NSC-92-2216-E-011-024, NSC-92-2622-E-002-014-cc3.

Contract grant sponsor: Ministry of Economic Affairs; contract grant number: 95-EC-17-A-11-S1-057.

Contract grant sponsor: Taiwan Textile Research Institute.

polymers were invented and filed as patents.<sup>3,5-9</sup> In our previous investigation,<sup>10</sup> the releasing properties of negative air ions of tourmaline containing polypropylene (PP) composite films under varying testing conditions were studied. It is interesting to note that the average concentrations of negative air ions ( $C_{\text{ion-}}$ ) emitted from PP/tourmaline composite specimens tested at varying conditions reached a maximum value as the tourmaline contents present in PP/tourmaline composite specimens approached the 5 wt % optimum value. Further morphology analysis of the PP/tourmaline composite specimens indicates that significant aggregation of tourmaline powders occur as their contents are higher than 5 wt %. Presumably, the significantly reduced  $C_{\text{ion-}}$  values of PP/tourmaline composite specimens with tourmaline contents more than 5 wt % is attributed to the aggregated tourmaline powders found at high tourmaline loadings, since the effective surface areas for generating negative air ions of tourmaline powders can be significantly reduced as the tourmaline powders are over-aggregated. Moreover, all  $C_{\text{ion-}}$  values of PP/tourmaline composite specimens tested at dynamic impact mode are significantly higher than those of the corresponding specimens tested at static mode but the same temperature. In fact, the  $C_{\text{ion-}}$  value of the P<sub>T5</sub> composite specimens tested at 90°C with dynamic impact mode of about 300 particles/cc is about 10 times more than that of pure PP film specimen tested at 25°C with static mode. In our recent investigation,<sup>11</sup> it was found that the  $C_{\text{ion-}}$  values of the blends of dynamically vulcanized ethylene-propylene-diene (EPDM)/PP/tourmaline tested at varying conditions are significantly higher than those of the PP/tourmaline specimens with the same loadings of tourmaline powders. A beneficial temperature and dynamic impact effect on  $C_{\text{ion-}}$  values of the EPDM/PP/tourmaline composite specimens was also found. The  $C_{\text{ion-}}$  values of the E/P<sub>T3</sub> composite specimens tested at 90°C with dynamic impact mode of about 620 particles/cc are about two times more than that of P<sub>T5</sub> film specimen tested at same condition. Presumably, the tourmaline-dispersed PP phases were not miscible and were expelled by the EPDM vulcanizates during the melt-blending processes of EPDM/PP/tourmaline resins. The tourmaline powders dispersed in PP phases and/or interfacial regions of EPDM/PP/tourmaline composite specimens are expected to confront with higher degrees of deformation and/or piezoelectric effect than those tourmaline particles dispersed in PP/tourmaline composite specimens with the same tourmaline loadings. The interesting negative air ion properties observed above are attributed to the pyroelectric and piezoelectric characteristics of the tourmaline powders present in PP/tourmaline and EPDM/PP/tourmaline composite specimens, respec-

tively, since the efficiency of generating negative air ions of the tourmaline crystals are expected to improve with the ambient temperatures or pressures.

Porous bamboo charcoals are well known for their excellent adsorption activity and high specific surface areas (ca. 200–300 m<sup>2</sup>/g)<sup>12</sup>. In addition to the fuel, they have been used as an adsorbent, catalyst support, humidity controller,<sup>13,14</sup> and wastewater purification substance.<sup>15,16</sup> Natural bamboo charcoals can generate negative air ions spontaneously and permanently without generating excess amounts of radiation or ozone molecules.<sup>17</sup> It is worth noting that the efficiency of generating negative air ions of the bamboo charcoal can be improved significantly by increasing their ambient temperatures or pressures, since they are generally recognized as pyroelectric and piezoelectric materials.<sup>17,18</sup> Most recently, bamboo charcoal containing polymers capable of emitting active ions and far-infrared lights were invented and filed as patents.<sup>16-18</sup> However, as far as we know, the active-ion emitting properties of bamboo charcoals containing polymers have never been reported in the literature.<sup>12-18</sup>

In this study, porous bamboo charcoals were used as the support to disperse tourmaline powders and to enhance the active-ion emitting properties of tourmaline/bamboo charcoal compounds containing PP or EPDM/PP composite specimens. The influences of compositions and contents of tourmaline/bamboo charcoal compounds on the active-ion emitting properties of tourmaline/bamboo charcoal compounds containing EPDM/PP composite specimens were investigated. The  $C_{\text{ion-}}$  values emitted from tourmaline/bamboo charcoal compounds containing PP and EPDM/PP composite specimens under “static” or “dynamic” impact conditions were investigated at varying temperatures. It is worth noting that  $C_{\text{ion-}}$  values of many tourmaline/bamboo charcoal compounds containing PP and EPDM/PP composite specimens are significantly higher than those of the corresponding composite specimens with the same loadings of tourmaline or bamboo charcoal powders, respectively. Possible reasons accounting for the enhanced active-ion emitting properties of tourmaline/bamboo charcoal compounds contained EPDM/PP composite specimens are proposed.

## EXPERIMENTAL

### Materials and sample preparation

The ethylene propylene diene terpolymer/polypropylene (EPDM/PP) blends were prepared via reactive extrusion, using a Nanjing Jiant SHJ-36 corotating twin screw extruder (Nanjing, China), wherein the length to diameter ratio of the screw is 40. The basic polymers used to prepare EPDM/PP

thermoplastic elastomers are PP and EPDM with ethylidene 2-norbonene (ENB) resins, which were obtained from Taiwan Polypropylene and Dupont Corporation, respectively. Dimethylol phenolic resin (SP 1405) and SnCl<sub>2</sub>, respectively, were obtained from Schenectady International and Mallinckrodt Baker, and were used as curing agents and accelerators to vulcanize the EPDM in the reactive extrusion processes, respectively. For every 60 parts by weight of EPDM, 40 parts of PP resins were melt-blended twice to shear and disperse the EPDM resins in the twin screw extruder, wherein the extruder was operated at 185°C in the feeding zone and at 190°C toward the extrusion die with a constant screw speed of 115 rpm. The pre-blended EPDM/PP resins were then quenched in cold water at 25°C and cut into pellet form. The pre-blended EPDM/PP resins prepared by pre-blending processes twice were then melt-blended with 10, 2, and 10 parts of dimethylol phenolic resin, SnCl<sub>2</sub>, and naphthenic process oil, respectively, for further dynamical vulcanization of the EPDM resin. During the dynamical vulcanization processes, the extruder was operated under the same conditions as those of the pre-blending process. The vulcanized EPDM/PP resins were then quenched in cold water at 25°C and cut into pellet form.

The tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal compounds filled EPDM/PP and/or PP master resins were first prepared by feeding the EPDM/PP and PP resins together with the fillers into the twin-screw extruder at a weight ratio of 80 : 20, respectively. Before the melt-blending processes of EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal master resins, the tourmaline powders were dry-blended with the bamboo charcoal particles thoroughly. To adhere the tourmaline/bamboo charcoal powders onto the surfaces of EPDM/PP and/or PP resins, 1 g of naphthalene oil was mixed with 100 g of the mixtures of EPDM/PP and/or PP resins with tourmaline/bamboo charcoal powders before compounding. The extruder was operated at 185°C in the feeding zone and at 195°C toward the extrusion die with a screw speed of 115 rpm. The extruded master resins were then quenched in cold water at 25°C and cut into the pellet form. The tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal containing EPDM/PP and/or PP resins were then prepared by melt-blending the corresponding master resins with varying amounts of pure EPDM/PP and/or PP resins in the twin-screw extruder, respectively, in which the same processing conditions mentioned above were used. Tables I and II summarize the compositions and specimen designations of the tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal compounds filled EPDM/PP and PP composite resins prepared in this study.

**TABLE I**  
**Compositions and Sample Designations of Tourmaline, Bamboo Charcoal, and Tourmaline/Bamboo Charcoal Filled PP Composite Resins**

Composition samples	Tourmaline (wt %)	Bamboo charcoal (wt %)	EPDM/PP (wt %)	PP (wt %)	
P <sub>(TB)1</sub>	P <sub>B1</sub>	0.0	1.0	0	99.0
	P <sub>T0.2B0.8</sub>	0.2	0.8	0	99.0
	P <sub>T0.4B0.6</sub>	0.4	0.6	0	99.0
	P <sub>T0.6B0.4</sub>	0.6	0.4	0	99.0
	P <sub>T0.8B0.2</sub>	0.8	0.2	0	99.0
P <sub>(TB)3</sub>	P <sub>T1</sub>	1.0	0.0	0	99.0
	P <sub>B3</sub>	0.0	3.0	0	97.0
	P <sub>T0.6B2.4</sub>	0.6	2.4	0	97.0
	P <sub>T1.2B1.8</sub>	1.2	1.8	0	97.0
	P <sub>T1.8B1.2</sub>	1.8	1.2	0	97.0
P <sub>(TB)4</sub>	P <sub>T2.4B0.6</sub>	2.4	0.6	0	97.0
	P <sub>T3</sub>	3.0	0.0	0	97.0
	P <sub>B4</sub>	0.0	4.0	0	96.0
	P <sub>T0.8B3.2</sub>	0.8	3.2	0	96.0
	P <sub>T1.6B2.4</sub>	1.6	2.4	0	96.0
P <sub>(TB)5</sub>	P <sub>T2.4B1.6</sub>	2.4	1.6	0	96.0
	P <sub>T3.2B0.8</sub>	3.2	0.8	0	96.0
	P <sub>T4</sub>	3.0	0.0	0	96.0
	P <sub>B5</sub>	0.0	5.0	0	95.0
	P <sub>T1B4</sub>	1.0	4.0	0	95.0
P <sub>(TB)10</sub>	P <sub>T2B3</sub>	2.0	3.0	0	95.0
	P <sub>T3B2</sub>	3.0	2.0	0	95.0
	P <sub>T4B1</sub>	4.0	1.0	0	95.0
	P <sub>T5</sub>	5.0	0.0	0	95.0
	P <sub>B10</sub>	0.0	10.0	0	90.0
	P <sub>T2B8</sub>	2.0	8.0	0	90.0
	P <sub>T4B6</sub>	4.0	6.0	0	90.0
	P <sub>T6B4</sub>	6.0	4.0	0	90.0
	P <sub>T8B2</sub>	8.0	2.0	0	90.0
	P <sub>T10</sub>	10.0	0.0	0	90.0

**Negative air ions analysis**

The composite specimens used for tensile and negative air ion analysis were prepared by hot-pressing the tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal compounds filled EPDM/PP and PP composite resins prepared above at 180°C for 5 min. The hot-pressed composite specimens were then cut into a dimension of 100 × 40 × 0.2 mm<sup>3</sup>, which were associated with a thickness of roughly 0.2 mm. The concentrations of negative air ions emitted from the composite specimens were determined using an Andes air ion detector model INTI ITC-201A, which was purchased from Andes electrical corporation, Qingsen, Japan. Before negative air ion analysis, the composite specimens were kept in a moisture regain box for 12 h, wherein the box was operated at 25°C and 40% relative humidity (RH). The concentrations of negative air ions emitted from the composite specimens were then determined at 45 ± 5% RH and at temperatures ranging from 25 to 90°C. The average concentrations of negative air ions determined in the first minute were then used as the

**TABLE II**  
**Compositions and Sample Designations of Tourmaline, Bamboo Charcoal, and Tourmaline/Bamboo Charcoal Filled EPDM/PP Composite Resins**

Compositions samples	Tourmaline (wt %)	Bamboo charcoal (wt %)	EPDM/PP (wt %)	PP (wt %)	
E/P <sub>(TB)1</sub>	E/P <sub>B1</sub>	0.0	1.0	99.0	0
	E/P <sub>T0.2B0.8</sub>	0.2	0.8	99.0	0
	E/P <sub>T0.4B0.6</sub>	0.4	0.6	99.0	0
	E/P <sub>T0.6B0.4</sub>	0.6	0.4	99.0	0
	E/P <sub>T0.8B0.2</sub>	0.8	0.2	99.0	0
E/P <sub>(TB)3</sub>	E/P <sub>T1</sub>	1.0	0.0	99.0	0
	E/P <sub>B3</sub>	0.0	3.0	97.0	0
	E/P <sub>T0.6B2.4</sub>	0.6	2.4	97.0	0
	E/P <sub>T1.2B1.8</sub>	1.2	1.8	97.0	0
	E/P <sub>T1.8B1.2</sub>	1.8	1.2	97.0	0
E/P <sub>(TB)4</sub>	E/P <sub>T2.4B0.6</sub>	2.4	0.6	97.0	0
	E/P <sub>T3</sub>	3.0	0.0	97.0	0
	E/P <sub>B4</sub>	0.0	4.0	0	96.0
	E/P <sub>T0.8B3.2</sub>	0.8	3.2	0	96.0
	E/P <sub>T1.6B2.4</sub>	1.6	2.4	0	96.0
E/P <sub>(TB)5</sub>	E/P <sub>T2.4B1.6</sub>	2.4	1.6	0	96.0
	E/P <sub>T3.2B0.8</sub>	3.2	0.8	0	96.0
	P <sub>T4</sub>	3.0	0.0	0	96.0
	E/P <sub>B5</sub>	0.0	5.0	95.0	0
	E/P <sub>T1B4</sub>	1.0	4.0	95.0	0
E/P <sub>(TB)10</sub>	E/P <sub>T2B3</sub>	2.0	3.0	95.0	0
	E/P <sub>T3B2</sub>	3.0	2.0	95.0	0
	E/P <sub>T4B1</sub>	4.0	1.0	95.0	0
	E/P <sub>T5</sub>	5.0	0.0	95.0	0
	E/P <sub>B10</sub>	0.0	10.0	90.0	0
E/P <sub>(TB)10</sub>	E/P <sub>T2B8</sub>	2.0	8.0	90.0	0
	E/P <sub>T4B6</sub>	4.0	6.0	90.0	0
	E/P <sub>T6B4</sub>	6.0	4.0	90.0	0
	E/P <sub>T8B2</sub>	8.0	2.0	90.0	0
	E/P <sub>T10</sub>	10.0	0.0	90.0	0

average concentrations of negative air ions emitted from the composite specimens. Two types of testing methods were performed to determine the average concentrations of negative air ions emitted from the sectioned composite specimens. In addition to the static method, the negative air ion concentrations of the composite specimens were also detected under a "dynamic impact" mode. The dimensions of the "dynamic impact" instrument were described in our previous investigations<sup>10,11</sup> and are shown in Figure 1. During the testing, the composite specimens were wrapped on the specimen holder and dynamically impacted by 500 g of loading at a frequency of 40 rpm. During the testing, the air ion detector was placed 10 cm above the composite specimens.

### Morphology and surface composition analysis of composite specimens

The composite specimens prepared in the previous section were observed using a HITACHI S-3000N scanning electron microscope (SEM) to examine the distribution of the filler particles in the composite

specimens. Before SEM examination, the composite specimens were etched using cyclohexane at 50°C for 24 h to remove the PP phases present on the fracture surfaces. The specimens were gold-coated at 15 keV for 30 s before SEM examinations. The particle sizes and distribution of the filler particles present in the composite specimens were also determined using an optical microscope equipped with a Winner 99 micro particle image processor, which was purchased from Jinan Winner Instruments Corporation, Jinan, China. The compositions on the surface of the composite specimens were determined with a Siemens D5000S energy dispersive X-rays (EDX) equipped on the scanning electron microscope described above.

### Tensile properties

The tensile properties of the composite specimens were determined using a Hungta Instron testing machine model HT-9112 at 25°C and a crosshead speed of 100 mm/min. The dimensions of the dog bone-shaped specimens were prepared according to ASTM D638 Type IV standard. A 35-mm gauge length was used during each tensile experiment. The values of tensile strength and elongation at break were obtained based on the average tensile results of at least five tensile specimens.

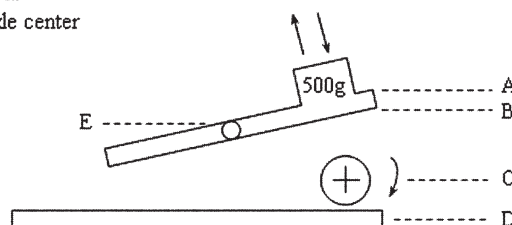
## RESULTS AND DISCUSSION

### Morphology and surface composition analysis

As shown in Figure 2, the average particle sizes of tourmaline or bamboo charcoal powders present in composite specimens increase slightly as their tourmaline or bamboo charcoal contents present in EPDM/PP and PP resins increase from 1 to 3 wt % and from 1 to 5 wt %, respectively. In contrast, the average particle sizes of the tourmaline or bamboo charcoal powders present in EPDM/PP and PP resins start to aggregate rapidly as their tourmaline or bamboo charcoal contents are higher than 3 or 5 wt

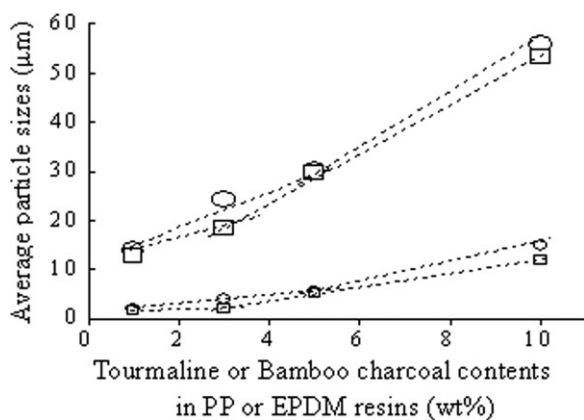
#### Dynamic impact instrument

- A: the loading
- B: the lever arm
- C: the heating roller
- D: the seat
- E: the axle center



**Figure 1** Illustration of the dynamic impact instrument.

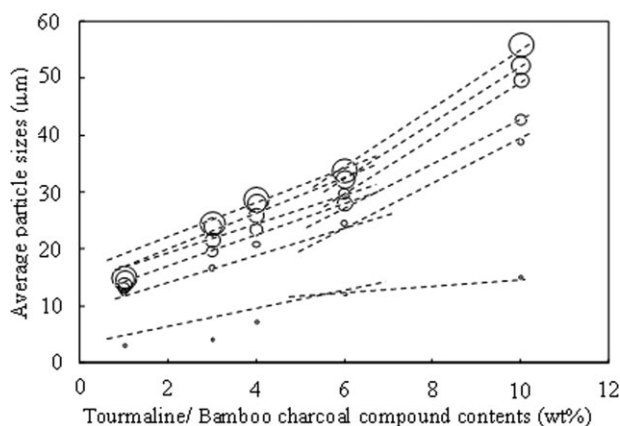




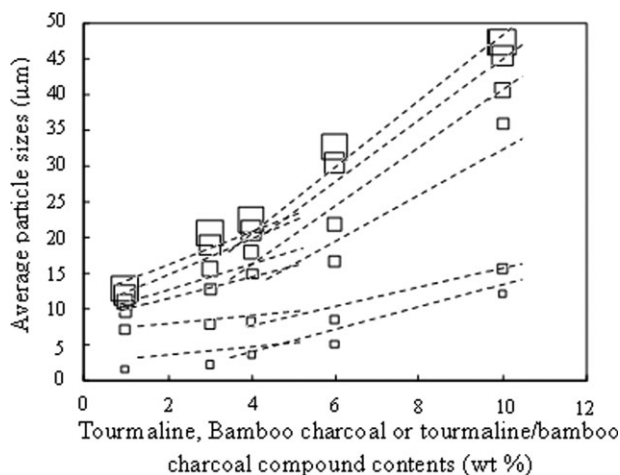
**Figure 2** The average particle sizes of tourmaline or bamboo charcoal powders present in  $P_B$  (○),  $EP_B$  (□),  $P_T$  (△), and  $EP_T$  (◇) film specimens evaluated using the particle image processor.

%, respectively. For convenience, the transition contents that correspond to the rapid coagulation of the tourmaline and bamboo charcoal powders in EPDM/PP or PP resins will be referred to as the “optimum contents” in the following discussion. As shown in Figure 2, the average particle sizes of tourmaline or bamboo charcoal powders containing EPDM/PP composite specimens increased rapidly from 2.1 to 12 µm and 18.5 to 53.3 µm, respectively, as their tourmaline or bamboo charcoal contents increased from the transition content of 3–10 wt %. Similarly, by increasing tourmaline or bamboo charcoal contents from the transition content of 5–10 wt %, the average particle sizes of tourmaline or bamboo charcoal powders containing PP composite specimens increased rapidly from 5 to 15 µm and 30 to 55.7 µm, respectively.

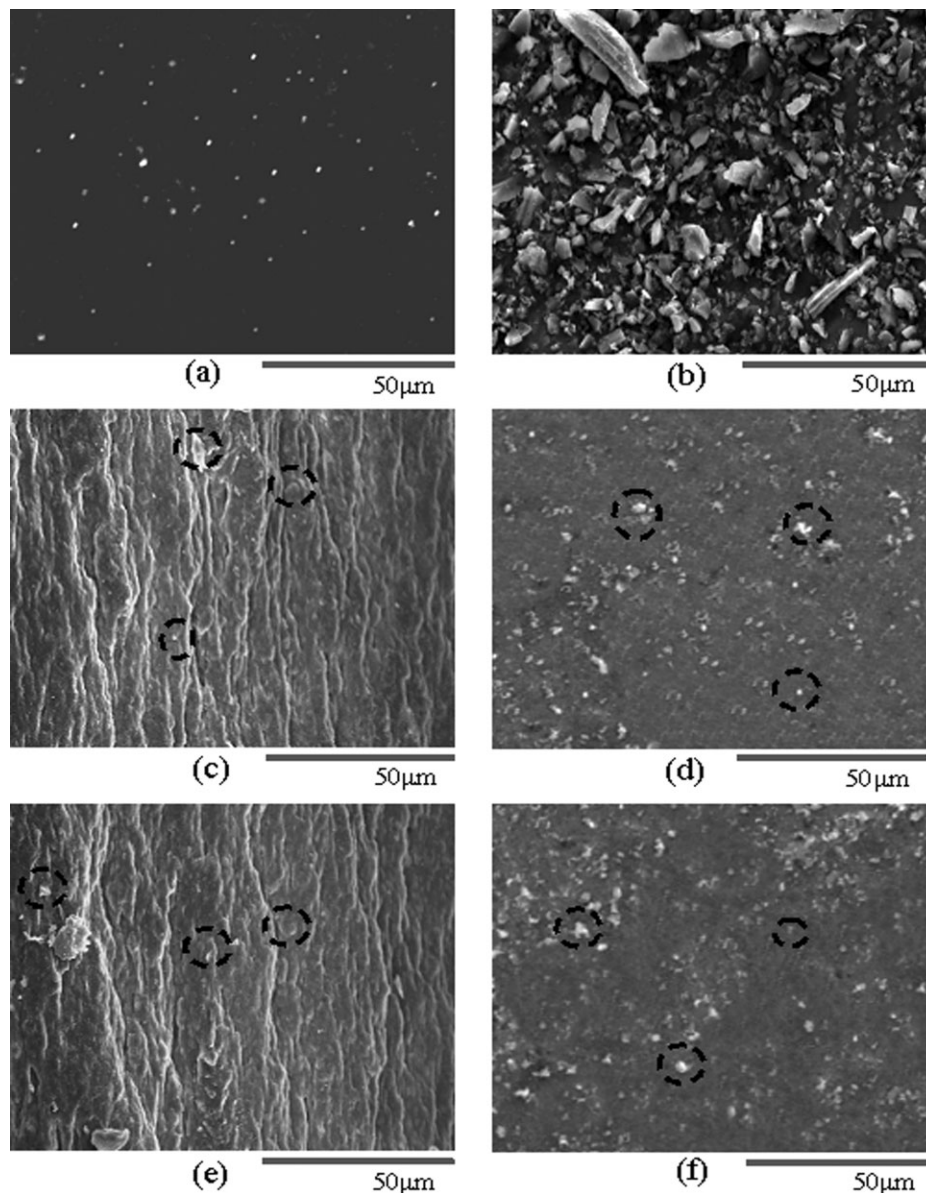
Figures 3 and 4 summarize the average particle sizes of tourmaline/bamboo charcoal compounds containing PP and EPDM/PP composite specimens, respectively. It is interesting to note that the “optimum contents” corresponding to the rapid coagulation of the tourmaline/bamboo charcoal compounds in each tourmaline/bamboo charcoal compounds containing EPDM/PP and PP composite specimens are 4 and 6 wt %, respectively, which are slightly larger than those of the tourmaline or bamboo charcoal containing EPDM/PP and PP composite specimens at 3 and 5 wt %, respectively. As shown in Figures 3 and 4, the average particle sizes of tourmaline/bamboo charcoal compounds increased significantly from 8.2 to 15.7 µm and 24.6 to 38.8 µm, as the contents of tourmaline/bamboo charcoal compounds of EPDM/PP/tourmaline/bamboo charcoal film and PP/tourmaline/bamboo charcoal specimens increased from 4 to 10 wt % and 6 to 10 wt %, respectively. These results clearly suggest that at tourmaline/bamboo charcoal compound contents



**Figure 3** The average particle sizes of tourmaline, bamboo charcoal, and/or tourmaline/bamboo charcoal compound powders present in  $P_{(TB)1}$  (i.e.  $P_{B1}$  (○),  $P_{T0.2B0.8}$  (○),  $P_{T0.4B0.6}$  (○),  $P_{T0.6B0.4}$  (○),  $P_{T0.8B0.2}$  (○) and  $P_{T1}$  (○)),  $P_{(TB)3}$  (i.e.  $P_{B3}$  (○),  $P_{T0.6B2.4}$  (○),  $P_{T1.2B1.8}$  (○),  $P_{T1.8B1.2}$  (○),  $P_{T2.4B0.6}$  (○) and  $P_{T3}$  (○)),  $P_{(TB)4}$  (i.e.  $P_{B4}$  (○),  $P_{T0.8B3.2}$  (○),  $P_{T1.6B2.4}$  (○),  $P_{T2.4B1.6}$  (○),  $P_{T3.2B0.8}$  (○) and  $P_{T4}$  (○)),  $P_{(TB)6}$  (i.e.  $P_{B6}$  (○),  $P_{T1.2B4.8}$  (○),  $P_{T2.4B3.6}$  (○),  $P_{T3.6B2.4}$  (○),  $P_{T4.8B1.2}$  (○) and  $P_{T6}$  (○)),  $P_{(TB)10}$  (i.e.  $P_{B10}$  (○),  $P_{T2B8}$  (○),  $P_{T4B6}$  (○),  $P_{T6B4}$  (○),  $P_{T8B2}$  (○), and  $P_{T10}$  (○)) series composite specimens evaluated using particle image processor.



**Figure 4** The average particle sizes of tourmaline, bamboo charcoal, or tourmaline/bamboo charcoal compound powders present in  $E/P_{(TB)1}$  (i.e.,  $E/P_{B1}$  (□),  $E/P_{T0.2B0.8}$  (□),  $E/P_{T0.4B0.6}$  (□),  $E/P_{T0.6B0.4}$  (□),  $E/P_{T0.8B0.2}$  (□) and  $E/P_{T1}$  (□)),  $E/P_{(TB)3}$  (i.e.  $E/P_{B3}$  (□),  $E/P_{T0.6B2.4}$  (□),  $E/P_{T1.2B1.8}$  (□),  $E/P_{T1.8B1.2}$  (□),  $E/P_{T2.4B0.6}$  (□) and  $E/P_{T3}$  (□)),  $E/P_{(TB)4}$  (i.e.  $E/P_{B4}$  (□),  $E/P_{T0.8B3.2}$  (□),  $E/P_{T1.6B2.4}$  (□),  $E/P_{T2.4B1.6}$  (□),  $E/P_{T3.2B0.8}$  (□) and  $E/P_{T4}$  (□)),  $E/P_{(TB)6}$  (i.e.  $E/P_{B6}$  (□),  $E/P_{T1.2B4.8}$  (□),  $E/P_{T2.4B3.6}$  (□),  $E/P_{T3.6B2.4}$  (□),  $E/P_{T4.8B1.2}$  (□) and  $E/P_{T6}$  (□)),  $E/P_{(TB)10}$  (i.e.  $E/P_{B10}$  (□),  $E/P_{T2B8}$  (□),  $E/P_{T4B6}$  (□),  $E/P_{T6B4}$  (□),  $E/P_{T8B2}$  (□), and  $E/P_{T10}$  (□)) series composite specimens evaluated using particle image processor.

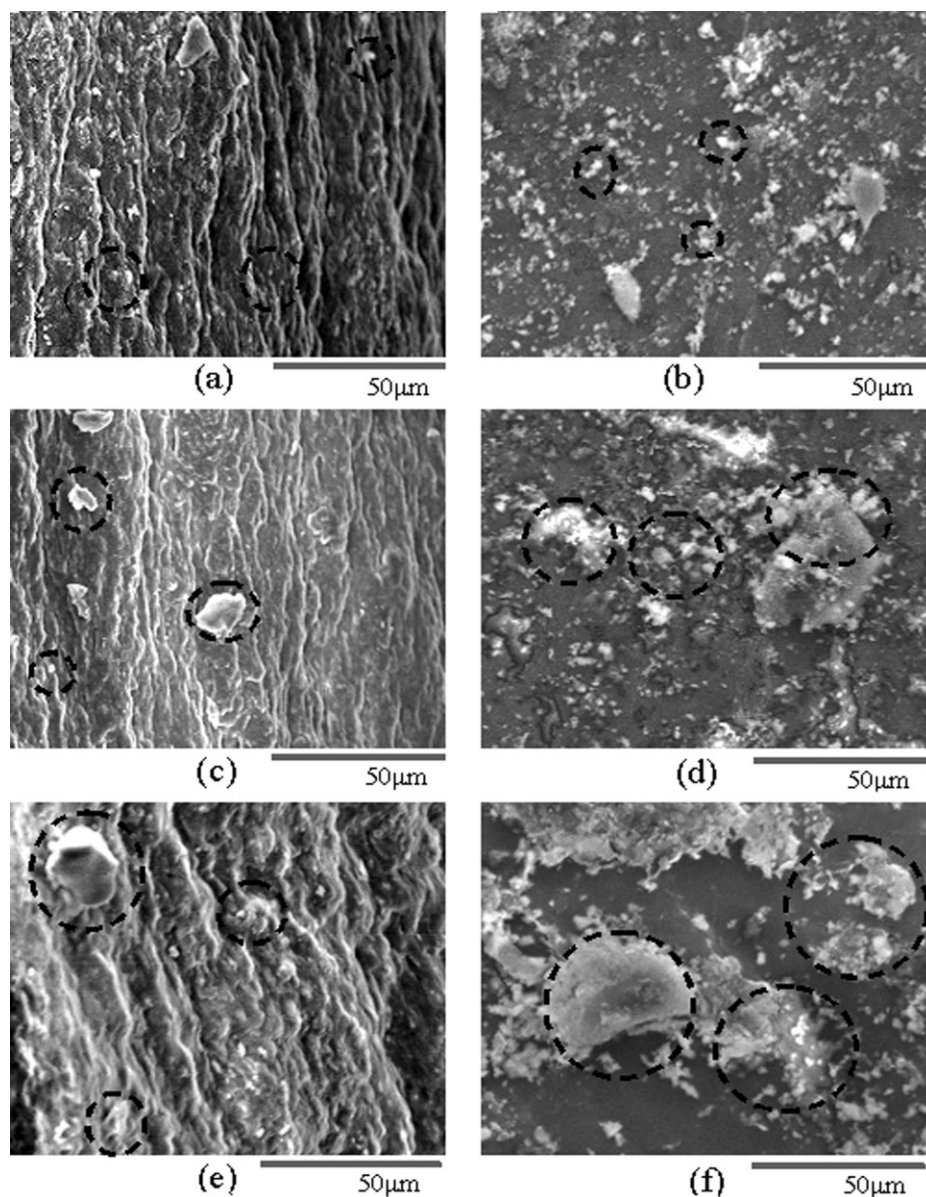


**Figure 5** SEM micrographs of the fracture surfaces of (a) pure tourmaline powders, (b) pure bamboo charcoals, (c) E/ $P_{T0.4B0.1}$ , (d)  $P_{T0.4B0.1}$ , (e) E/ $P_{T0.8B0.2}$ , and (f)  $P_{T0.8B0.2}$  specimens. (The dotted lines circulate the tourmaline and bamboo charcoal particles distributed in the specimens.)

lower than 4 and 6 wt %, the tourmaline/bamboo charcoal compound powders can be relatively well dispersed in EPDM/PP or PP matrices by melt blending; however, significant aggregation of the tourmaline/bamboo charcoal compound powders begin as their contents are higher than their corresponding "transition contents."

Typical SEM micrographs of tourmaline, bamboo charcoal powders, and the fracture surfaces of EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal composite specimens are summarized in Figures 5(a–f) and 6(a–f), respectively. As shown in Figure 5(a), the average particle size of the original tourmaline powders is about 0.5  $\mu\text{m}$ , with relatively small size distribution. In com-

parison with the tourmaline powders, the bamboo charcoal powders exhibit larger average particle size (ca. 5.3  $\mu\text{m}$ ), aspect ratio, and size distribution [see Fig. 5(b)]. However, some of the tourmaline/bamboo charcoal compound powders coagulated significantly during the preparation processes of EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal composite specimens [see Figs. 5(c–f) and 6(a–f)]. Figures 7 and 8 summarize typical enlarged micrographs and EDX analysis of the compositions of tourmaline/bamboo charcoal compound particles on the surfaces of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens present in Figures 7(a–d) and 8(a–d), respectively. It is



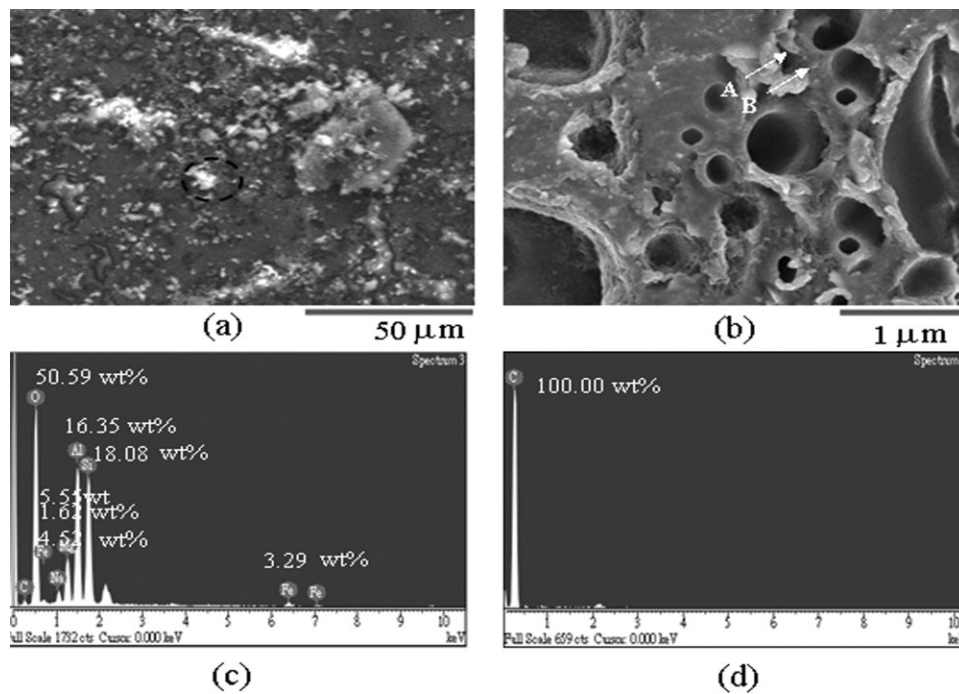
**Figure 6** SEM micrographs of the fracture surfaces of (a) E/P<sub>T2.4B0.6</sub>, (b) P<sub>T2.4B0.6</sub>, (c) E/P<sub>T4.8B1.2</sub>, (d) P<sub>T4.8B1.2</sub>, (e) E/P<sub>T8B2</sub>, and (f) P<sub>T8B2</sub> specimens. (The dotted lines circulate the tourmaline and bamboo charcoal particles distributed in the specimens.)

interesting to note that the bamboo charcoal powders are highly porous substances, wherein pore cavities with sizes ranging from 0.2 to several  $\mu\text{m}$  were found distributed in the bamboo charcoal powders. As shown in Figures 7(b) and 8(b), the particles located on position "A" of the surfaces of bamboo charcoal particles of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens are composed of O, Al, Si, Fe, C, and Na elements, respectively. These compositions are very similar to those of the plain tourmaline powders,<sup>10–12</sup> which are composed of more than 98 wt % of O, Al, Si, and Fe elements. Apparently, many of the relatively small tourmaline powders were dispersed in the cavities of bamboo charcoal

particles during the preparation processes of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens.

It is not clear what accounts for these interesting morphology properties observed for PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal specimens. However, it is generally recognized that the tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal compound powders can only be dispersed into PP matrices of EPDM/PP specimens, since the vulcanized EPDM phase cannot be melted and/or be barely melted again during the melt-blending processes of tourmaline/bamboo charcoal powders and EPDM/PP resins. Under such circumstances, tourmaline, bamboo

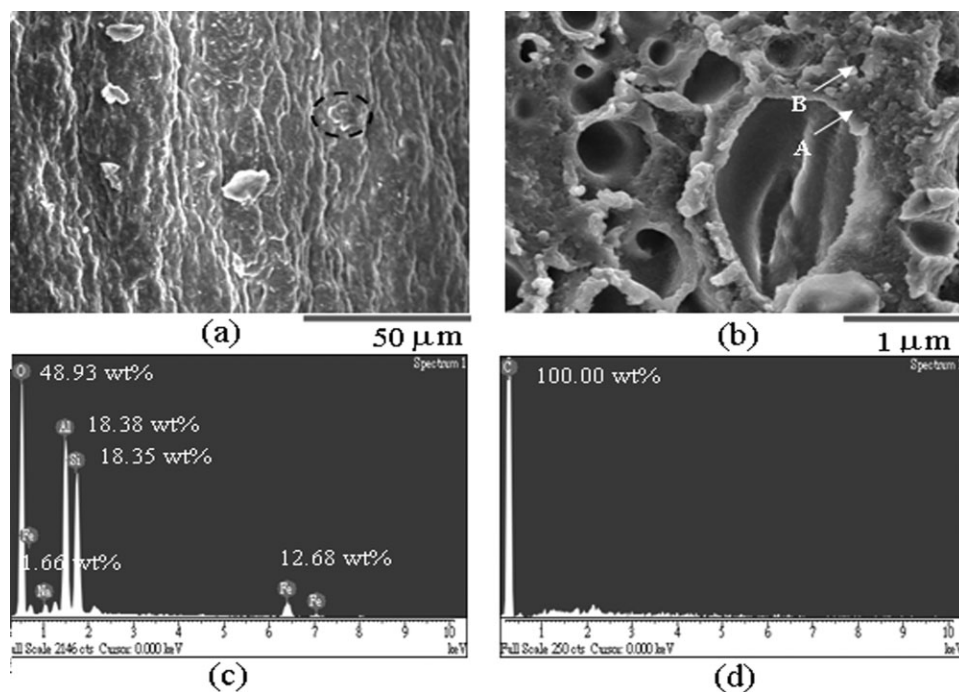




**Figure 7** SEM micrographs of the fracture surfaces of (a)  $P_{T4.8B1.2}$  film specimen, (b) the high-magnification micrograph of circled section in (a), EDX showing spectrum on elemental composition information of the tourmaline and bamboo charcoal compounds present in (c) A and (d) B position of (b), respectively.

charcoal, and tourmaline/bamboo charcoal compound powders are mostly expelled by EPDM vulcanizes into the melting fronts of PP and dispersed in PP and/or the interfacial areas between EPDM

and PP matrices during the melt-blending processes. The amounts of tourmaline or bamboo charcoal powders needed to be well dispersed in PP matrices of EPDM/PP/tourmaline or EPDM/PP/bamboo



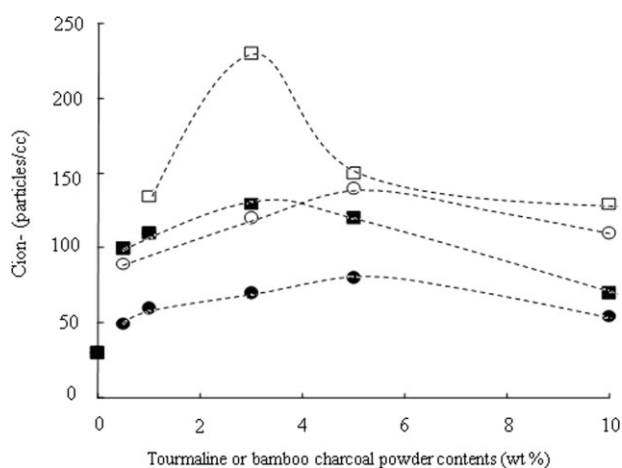
**Figure 8** SEM micrographs of the fracture surfaces of (a) E/ $P_{T4.8B1.2}$  film specimen, (b) the high-magnification micrograph of circled section in (a), EDX showing spectrum on elemental composition information of the tourmaline and bamboo charcoal compounds present in (c) A and (d) B position of (b), respectively.



charcoal specimens are expected to be significantly smaller than those needed in PP/tourmaline or PP/bamboo charcoal specimens. Based on these premises, it is reasonable to believe that the optimum tourmaline or bamboo charcoal contents of EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal specimens is significantly lower than that of the PP/tourmaline and PP/bamboo charcoal specimens (i.e., 3 and 5 wt %). On the other hand, as evidenced by the SEM morphological studies, the dispersed tourmaline particles found in the cavities of bamboo charcoal particles is expected to increase the optimum contents of tourmaline/bamboo charcoal compounds in PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens before rapid coagulation of the tourmaline/bamboo charcoal compound powders occurs. It is, therefore, the optimum loading contents of tourmaline/bamboo charcoal compound increase to 4 and 6 wt % for EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal specimens, respectively.

### Negative air ion releasing properties

Figure 9 summarizes the average concentrations of negative air ions ( $C_{ion-}$ ) emitted from tourmaline or bamboo charcoal contained PP and EPDM/PP composite specimens tested at 25°C and under static mode, respectively. As shown in Figure 9, about 30 particles/cc of negative air ions were emitted from the pure PP and EPDM/PP film specimen tested at static mode and 25°C, respectively. After blending tourmaline or bamboo charcoal powders in PP resins, the  $C_{ion-}$  values of PP/tourmaline or PP/bamboo charcoal composite specimens increase sig-



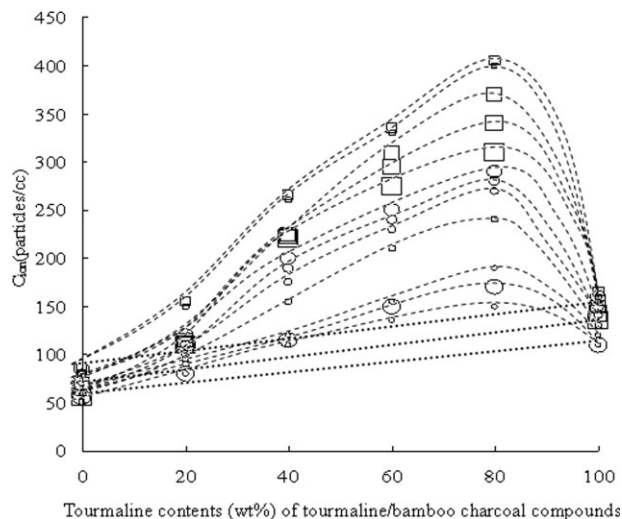
**Figure 9** Average concentrations of negative air ions ( $C_{ion-}$ ) emitted From  $P_T$  (○),  $P_B$  (●),  $E/P_T$  (□), and  $E/P_B$  (■) composite specimens with varying contents of tourmaline powders at 25°C.

nificantly with the increasing tourmaline or bamboo charcoal contents, respectively. The  $C_{ion-}$  values of PP/tourmaline or PP/bamboo charcoal composite specimens reach the maximum as their tourmaline or bamboo charcoal contents approach the optimum value of 5 wt %. It is interesting to note that, at the same loading of tourmaline or bamboo charcoal powders, the  $C_{ion-}$  values of PP/bamboo charcoal composite specimens are always significantly lower than those of the PP/tourmaline composite specimens. For instance, the  $C_{ion-}$  values of the PP/tourmaline and PP/bamboo charcoal specimens with 5 wt % optimum filler loading of tourmaline (i.e.,  $P_{T5}$  and  $P_{B5}$ ) are 140 and 80 particles/cc, respectively. Similar negative air ion releasing properties were found in EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal composite specimens, wherein the  $C_{ion-}$  values of EPDM/PP/bamboo charcoal composite specimens are significantly lower than those of the EPDM/PP/tourmaline composite specimens with the same loadings of tourmaline or bamboo charcoal powders. As evidenced from previous morphological investigation, significant aggregation of the tourmaline or bamboo charcoal powders start to occur as their contents are higher than their corresponding optimum contents, respectively. These significantly aggregated tourmaline or bamboo charcoal powders are expected to reduce their effective surface areas for emitting negative air ions at high tourmaline or bamboo charcoal loadings, respectively. As a consequence, PP/tourmaline or PP/bamboo charcoal composite specimens exhibit the maximum  $C_{ion-}$  values as their tourmaline or bamboo charcoal contents approach the optimum value of 5 wt %.

On the other hand, it is worth noting that the  $C_{ion-}$  values of EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal composite specimens are always higher than those of the corresponding PP/tourmaline and PP/bamboo charcoal composite specimens with the same loadings of tourmaline or bamboo charcoal powders, respectively (see Fig. 9). In contrast to the 5 wt % optimum loading found in PP/tourmaline and/or PP/bamboo charcoal composite specimens, EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal composite specimens exhibit the maximum  $C_{ion-}$  values as their tourmaline or bamboo charcoal contents approach the optimum value of 3 wt %. The  $C_{ion-}$  values of the EPDM/PP/tourmaline and EPDM/PP/tourmaline specimens with 3 wt % optimum loading of tourmaline (i.e.,  $E/P_{T3}$  and  $E/P_{B3}$ ) reach about 230 and 130 particles/cc, respectively, which are about 7 and 4 times more than those of the pure PP and/or EPDM/PP specimens tested under static mode and at 25°C, respectively. At tourmaline or bamboo charcoal loadings higher than their corresponding optimum values, the  $C_{ion-}$  values of PP/tourmaline,

PP/bamboo charcoal, EPDM/PP/tourmaline, and EPDM/PP/bamboo charcoal composite specimens reduce significantly with further increase in tourmaline or bamboo charcoal contents, respectively. As suggested in our previous investigation,<sup>11</sup> the tourmaline and/or bamboo charcoal dispersed PP phases are expected to be not miscible and are expelled by the EPDM vulcanizates during the melt-blending processes of EPDM/PP/tourmaline resins. Certain degrees of residual strains can be present between the interfacial regions of tourmaline or bamboo charcoal dispersed PP and/or EPDM phases. Under such circumstances, the tourmaline or bamboo charcoal powders dispersed in PP phases of EPDM/PP/tourmaline composite specimens are expected to confront with higher degrees of deformation and/or piezoelectric effect than those tourmaline and/or bamboo charcoal compound particles dispersed in PP/tourmaline composite specimens with the same loadings. Based on these premises, it is reasonable to believe that, at the same testing condition, the  $C_{ion-}$  values of EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal specimens are always higher than those of the corresponding PP/tourmaline and PP/bamboo charcoal specimens with the same loadings of tourmaline powders, respectively.

Figure 10 summarizes the average concentrations of negative air ions ( $C_{ion-}$ ) emitted from tourmaline/bamboo charcoal compounds contained PP and EPDM/PP composite specimens tested at 25°C and under static mode, respectively. It is interesting to note that the  $C_{ion-}$  values of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens are significantly higher than their corresponding theoretical  $C_{ion-}$  values estimated using the "simple mixing rule." Moreover, at a fixed content of the tourmaline/bamboo charcoal compound, the  $C_{ion-}$  values of PP/tourmaline/bamboo charcoal composite specimens reach a maximum value as the weight ratio of tourmaline to bamboo charcoal of the tourmaline/bamboo charcoal compounds reaches 4. In fact, the  $C_{ion-}$  values of PP/tourmaline/bamboo charcoal composite specimens with the optimum tourmaline/bamboo charcoal weight ratio of 4 reach another maximum as their total contents of tourmaline/bamboo charcoal compounds reach the optimum loading of 6 wt % (i.e.,  $P_{T4.8B1.2}$  specimen). As shown in Figure 10, the  $C_{ion-}$  value of  $P_{T4.8B1.2}$  specimen reach about 270, which is about eight times more than that of the pure PP and EPDM/PP specimens tested under static mode and at 25°C. Similarly, the  $C_{ion-}$  values of EPDM/PP/tourmaline/bamboo charcoal composite specimens with the optimum tourmaline/bamboo charcoal weight ratio of 4 also reach another maximum as their total contents of tourmaline/bamboo



**Figure 10** The average concentrations of negative air ions ( $C_{ion-}$ ) emitted from  $P_{(TB)1}$  (○),  $P_{(TB)3}$  (○),  $P_{(TB)4}$  (○),  $P_{(TB)5}$  (○),  $P_{(TB)6}$  (○),  $P_{(TB)10}$  (○) and  $E/P_{(TB)1}$  (□),  $E/P_{(TB)3}$  (□),  $E/P_{(TB)4}$  (□),  $E/P_{(TB)5}$  (□),  $E/P_{(TB)6}$  (□),  $E/P_{(TB)10}$  (□) film series specimens with varying contents of tourmaline/bamboo charcoal compounds at 25°C. (The dashed line represents the theoretical  $C_{ion-}$  values of the above tourmaline/bamboo charcoal compounds contained PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal film specimen calculated using simple mixing rule.)

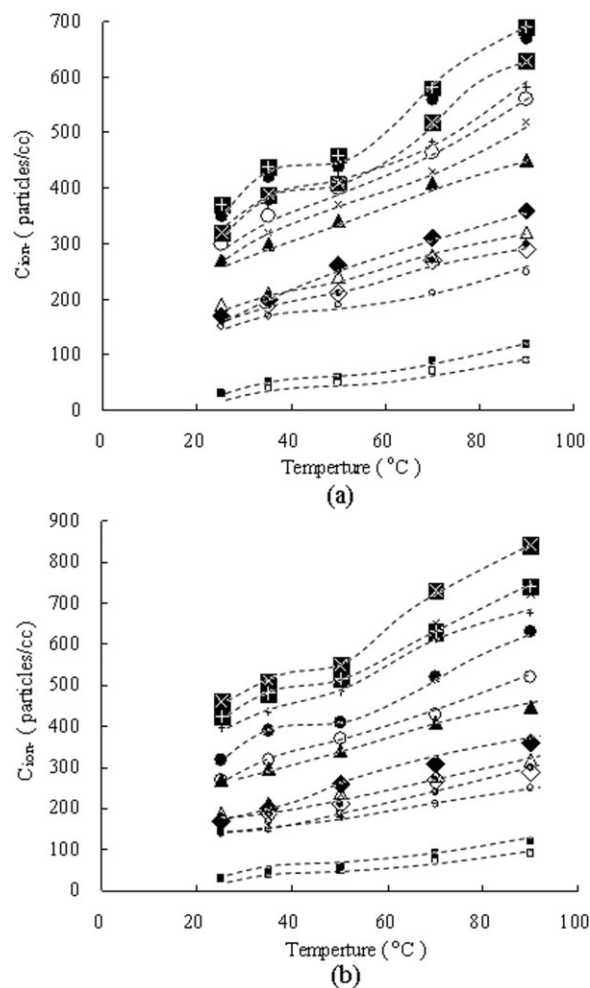
charcoal compounds reach the optimum loading of 4 wt %. However, it is worth noting that the  $C_{ion-}$  values of EPDM/PP/tourmaline/bamboo charcoal specimens are significantly higher than those of the PP/tourmaline/bamboo charcoal specimens with the same loadings of tourmaline/bamboo charcoal compounds. As shown in Figure 10, the  $C_{ion-}$  value of  $E/P_{T3.2B0.8}$  specimen reaches about 405, which is 13 times more than that of the pure PP and EPDM/PP specimens tested under static mode and at 25°C. As evidenced from previous SEM morphological investigations, many tourmaline powders were found in the cavities of bamboo charcoal particles of EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal specimens. The optimum contents and effective surface areas for emitting negative air ions of tourmaline/bamboo charcoal compounds are expected to increase significantly because of the well-dispersed tourmaline powders found in the cavities of bamboo charcoal particles of tourmaline/bamboo charcoal compounds. In comparison with the tourmaline or bamboo charcoal filled PP and/or EPDM/PP specimens, the "synergistically" improved releasing properties of negative air ions of the EPDM/PP/tourmaline/bamboo charcoal and PP/tourmaline/bamboo charcoal specimens due to the dispersed tourmaline powders present in

the cavities of bamboo charcoal particles of the tourmaline/bamboo charcoal compounds. However, it is not clear what accounts for the best negative air ion releasing properties always observed for the PP/tourmaline/bamboo charcoal and/or EPDM/PP/tourmaline/bamboo charcoal composite specimens with the optimum tourmaline to bamboo charcoal weight ratio at 4.

The  $C_{ion-}$  values of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens tested at varying temperatures and testing modes (i.e., static and dynamic impact) are summarized in Figure 11(a,b). The  $C_{ion-}$  values increase significantly as the testing temperatures of each PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal film specimen increased from 25 to 95°C. For example, the  $C_{ion-}$  values of  $P_{T4.8B1.2}$  and  $E/P_{T3.2B0.8}$  film specimen tested at static mode increase from 120 to 290 particles/cc and 155 to 405 particles/cc as the temperatures increased from 25 to 90°C. On the other hand, it is interesting to note that the  $C_{ion-}$  values of all PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens tested at dynamic impact mode were significantly higher than those of the same specimens tested at static mode but the same temperature [see Fig. 11(b)]. For instance, the  $C_{ion-}$  values of  $P_{T4.8B1.2}$  and  $E/P_{T3.2B0.8}$  film specimens tested at dynamic impact mode and 90°C reach near 690 and 840 particles/cc, respectively, which are about 100% more than those of the corresponding specimens tested at static mode and 25°C. Presumably, these interesting negative air ion properties of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens are attributed to the pyroelectric and piezoelectric characteristics of the tourmaline/bamboo charcoal powders present in the composite specimens, since the efficiency of releasing negative air ions of the tourmaline/bamboo charcoal crystals is expected to improve with the ambient temperatures or pressures.

### Tensile properties

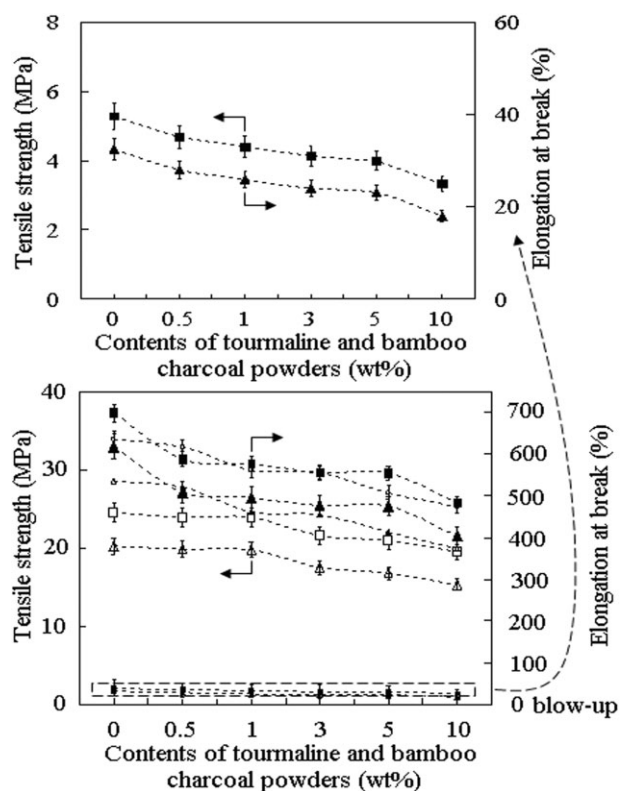
Figure 12 displays the tensile properties of tourmaline or bamboo charcoal filled PP and EPDM/PP composite specimens, respectively. As expected, the values of tensile strengths ( $\sigma_f$ ) and elongation at break ( $\epsilon_f$ ) of PP/tourmaline, PP/bamboo charcoal, EPDM/PP/tourmaline, and EPDM/PP/bamboo charcoal specimens reduce significantly as their tourmaline or bamboo charcoal powder contents increase. At the same tourmaline or bamboo charcoal loadings, EPDM/PP/tourmaline and EPDM/PP/bamboo charcoal composite specimens exhibit significantly higher  $\epsilon_f$  but slightly lower  $\sigma_f$  values



**Figure 11** The average concentrations of negative air ions ( $C_{ion-}$ ) emitted from (a) P ( $\square$ ,  $\blacksquare$ ),  $P_{T0.8B0.2}$  ( $\circ$ ,  $\bullet$ ),  $P_{T2.4B0.6}$  ( $\triangle$ ,  $\blacktriangle$ ),  $P_{T3.2B0.8}$  ( $\times$ ,  $\blacksquare$ ),  $P_{T4B1}$  ( $\diamond$ ,  $\blacklozenge$ ),  $P_{T4.8B1.2}$  ( $+$ ,  $\blacksquare$ ),  $P_{T8B2}$  ( $\diamond$ ,  $\blacklozenge$ ) and (b) E/P ( $\square$ ,  $\blacksquare$ ), E/ $P_{T0.8B0.2}$  ( $\circ$ ,  $\bullet$ ), E/ $P_{T2.4B0.6}$  ( $\triangle$ ,  $\blacktriangle$ ), E/ $P_{T3.2B0.8}$  ( $\times$ ,  $\blacksquare$ ), E/ $P_{T4B1}$  ( $\circ$ ,  $\bullet$ ), E/ $P_{T4.8B1.2}$  ( $+$ ,  $\blacksquare$ ) and E/ $P_{T8B2}$  ( $\diamond$ ,  $\blacklozenge$ ) composite film specimens tested under static and dynamic impact modes and at varying temperatures. (Hollow and solid symbols represent specimens tested under static and dynamic impact modes, respectively.)

than the corresponding PP/tourmaline and PP/bamboo charcoal composite specimens, respectively. As shown in Figure 12, the  $\epsilon_f$  values of EPDM/PP/tourmaline composite specimens reduce from 589.0 to 484.5% as their tourmaline contents increase from 0.5 to 10 wt %, respectively. However, the  $\epsilon_f$  values of PP/tourmaline composite specimens are only 35.0 and 25.0% as their tourmaline contents reach 0.5 and 10 wt %, respectively. The presence of EPDM phases can significantly improve the extensibility of the composite specimens, since the EPDM phases can be easily deformed under relatively low loadings. Under such circumstance, the filler particles present in the interfacial regions of EPDM/PP phases are





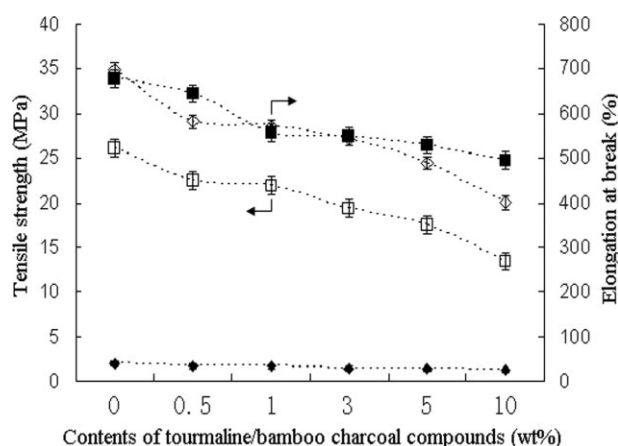
**Figure 12** The values of tensile strength ( $\square$ ,  $\triangle$ ) and elongation at break ( $\blacksquare$ ,  $\blacktriangle$ ) of PP/tourmaline, PP/bamboo charcoal and EPDM/PP/tourmaline, and EPDM/PP/bamboo charcoal composite specimens, respectively. (Large hollow and solid symbols represent the tensile strength and elongation at break of EPDM/PP/tourmaline or EPDM/PP/bamboo charcoal composite specimens, respectively. Symbol (I) represents the error bars of the tensile tests.)

likely deformed to higher degrees than those present in the PP/tourmaline specimens. Under such circumstance, the filler particles present in the interfacial regions of EPDM/PP phases are likely deformed to higher degrees than those present in the PP/tourmaline composite specimens. It is, therefore, reasonable to infer that filler particles filled EPDM/PP specimens can emit higher  $C_{ion-}$  values than those filler particle filled PP specimens with the same tourmaline or bamboo charcoal content and tested at the same condition. On the other hand, it is worth noting that the  $\sigma_f$  and  $\varepsilon_f$  values of tourmaline filled composite specimens (i.e., PP/tourmaline and EPDM/PP/tourmaline) are significantly higher than those of the bamboo charcoal filled composite specimens (i.e., PP/bamboo and EPDM/PP/bamboo charcoal) with the same filler loadings, respectively. As shown in Figure 12, the  $\sigma_f$  and  $\varepsilon_f$  values of EPDM/PP/tourmaline composite specimens are about 20 and 500% higher than those of the corre-

sponding EPDM/PP/bamboo charcoal composite specimens with the same filler loadings, respectively.

The tensile properties of typical tourmaline/bamboo charcoal compounds filled PP and EPDM/PP composite specimens are summarized in Figure 13. Similar to those found in the tourmaline or bamboo charcoal filled PP or EPDM/PP composite specimens, the  $\sigma_f$  and  $\varepsilon_f$  values of the tourmaline/bamboo charcoal compounds filled PP and EPDM/PP composite specimens reduce significantly with increasing loadings of the tourmaline/bamboo charcoal compounds, wherein the compounds are with the optimum tourmaline/bamboo charcoal weight ratio of 4. However, it is worth noting that PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens exhibit significantly higher  $\sigma_f$  and  $\varepsilon_f$  values than those of the PP/bamboo charcoal and EPDM/PP/bamboo charcoal composite specimens with the same filler loadings, respectively. For instance, at a filler loading of 3 wt %, the  $\sigma_f$  and  $\varepsilon_f$  values of P<sub>T2.4B0.6</sub> and E/P<sub>T2.4B0.6</sub> specimens are 27.4 MPa/29.7% and 19.3 MPa/543.7%, which yield about 112%/124% and 110%/114% higher than those of the P<sub>B3</sub> and E/P<sub>B3</sub> specimens, respectively.

Presumably, during the tensile experiments of these tourmaline, bamboo charcoal or tourmaline/bamboo charcoal compounds filled PP or EPDM/PP specimens, the presence of the filler particles can cause "stress concentration" and "early breakage of EPDM/PP molecules" at the boundaries between filler particles and the EPDM/PP resins, since the



**Figure 13** The values of tensile strength ( $\square$ ,  $\diamond$ ) and elongation at break ( $\blacksquare$ ,  $\blacklozenge$ ) of PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens. (Large hollow and solid symbols represent the tensile strength and elongation at break of EPDM/PP/tourmaline/bamboo charcoal composite specimens, respectively. Symbol (I) represents the error bars of the tensile tests.)

interfacial adhesion between the filler particles and EPDM/PP resins is poor. The degree of stress concentration is expected to increase as the filler loadings increase. It is, therefore, reasonable to infer that the  $\sigma_f$  and  $\varepsilon_f$  values of the tourmaline, bamboo charcoal, or tourmaline/bamboo charcoal compounds filled PP or EPDM/PP samples continue to reduce with increasing the filler loadings. However, the density of the tourmaline powders is much higher than that of the bamboo charcoal particle (3.1 g/cm<sup>3</sup> vs. 0.91 g/cm<sup>3</sup>). The volumes of the tourmaline powders are much smaller than those of the bamboo charcoal particles with the same weights. The degree of "stress concentration" and "early breakage of PP and/or EPDM molecules" at the boundaries is expected to reduce as the volumes of the filler particles reduce. On the other hand, the relatively large bamboo charcoal powders (c.a. 5.3  $\mu\text{m}$ ) are likely to cause easier stress concentration on the interfacial regions than those between polymer matrices and tourmaline powders with significantly lower particle sizes (c.a. 0.5  $\mu\text{m}$ ). As a consequence, PP/tourmaline and EPDM/PP/tourmaline samples always exhibit higher  $\sigma_f$  and  $\varepsilon_f$  values than the PP/bamboo charcoal and EPDM/PP/bamboo charcoal samples with the same filler loadings, respectively. However, the apparent loadings of the tourmaline/bamboo charcoal compounds present in PP/tourmaline/bamboo charcoal or EPDM/PP/tourmaline/bamboo charcoal specimens are expected to reduce significantly since many tourmaline powders can be dispersed in the cavities of the bamboo charcoal particles rather than distributing in the PP matrices. Based on these premises, it is reasonable to infer that the  $\sigma_f$  and  $\varepsilon_f$  values of the tourmaline/bamboo charcoal compounds filled PP and EPDM/PP specimens are always significantly higher than those of the tourmaline or bamboo charcoal filled PP and EPDM/PP specimens with the same filler loadings, respectively.

## CONCLUSIONS

The efficiency of generating negative air ions of PP/tourmaline, PP/bamboo charcoal, and EPDM/PP/tourmaline EPDM/PP/bamboo charcoal composite specimens improves initially as the filler loadings increase, although the  $C_{\text{ion-}}$  values of bamboo charcoal filled specimens are significantly lower than those of the corresponding tourmaline filled composite specimens with the same filler loadings. The optimum filler contents corresponding to the maximum  $C_{\text{ion-}}$  value of E/P<sub>T3</sub> and E/P<sub>B3</sub> specimens is 3 wt %, which is significantly lower than 5 wt % of the optimum filler content of P<sub>T5</sub> and P<sub>B5</sub> specimens. It is worth noting that the  $C_{\text{ion-}}$  values of tourmaline/

bamboo charcoal compounds filled PP and EPDM/PP composite specimens are significantly higher than their corresponding theoretical  $C_{\text{ion-}}$  values estimated using the "simple mixing rule." In fact, at a fixed tourmaline/bamboo charcoal loading, the  $C_{\text{ion-}}$  values of tourmaline/bamboo charcoal compounds filled PP or EPDM/PP composite specimens reach a maximum value as the weight ratio of tourmaline to bamboo charcoal of the tourmaline/bamboo charcoal compounds reaches 4. At this optimum tourmaline/bamboo charcoal weight ratio, the  $C_{\text{ion-}}$  values of tourmaline/bamboo charcoal compounds filled PP and EPDM/PP composite specimens reach another maximum as their total compound loadings reach the optimum loading of 6 and 4 wt % (i.e., P<sub>T4.8B1.2</sub>, and E/P<sub>T3.2B0.8</sub> specimens), respectively. The  $C_{\text{ion-}}$  values of the PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens increase significantly as they were tested under dynamic mode or with increasing the testing temperatures. For instance, the  $C_{\text{ion-}}$  values of P<sub>T5B1</sub> and E/P<sub>T3.2B0.8</sub> film specimens tested at dynamic impact mode and 90°C reach near 690 and 810 particles/cc, respectively, which are about 23 and 27 times more than that of the pure PP and EPDM/PP specimens tested under static mode and 25°C. Presumably, the significantly reduced  $C_{\text{ion-}}$  values of tourmaline, bamboo charcoal, and tourmaline/bamboo charcoal compounds filled PP and EPDM/PP composite specimens with filler contents more than their corresponding optimum loadings are attributed to the aggregated filler particles found at high filler loadings, since the effective surface areas for generating negative air ions of tourmaline, bamboo charcoal and tourmaline/bamboo charcoal compound particles can be significantly reduced as the fillers are over-aggregated. The significantly increased optimum contents and improved  $C_{\text{ion-}}$  values of tourmaline/bamboo charcoal compounds filled PP and EPDM/PP composite specimens are attributed to the well-dispersed tourmaline powders found in the cavities of bamboo charcoal particles of tourmaline/bamboo charcoal compounds.

The  $\sigma_f$  and  $\varepsilon_f$  values of tourmaline, bamboo charcoal, or tourmaline/bamboo charcoal compound particles filled PP and EPDM/PP composite specimens reduce significantly as their filler contents increase, respectively. However, the tourmaline filled PP and EPDM/PP composite specimens exhibit significantly higher  $\sigma_f$  and  $\varepsilon_f$  values than those of the bamboo charcoal filled PP and EPDM/PP specimens with the same filler loadings, respectively. Moreover, PP/tourmaline/bamboo charcoal and EPDM/PP/tourmaline/bamboo charcoal composite specimens exhibit significantly higher  $\sigma_f$  and  $\varepsilon_f$  values than those of the PP/bamboo charcoal and EPDM/PP/bamboo charcoal composite specimens with the

same filler loadings, respectively. Presumably, this is because the volumes of the tourmaline powders and/or tourmaline dispersed bamboo charcoal compounds are much smaller than those of the pure bamboo charcoal particles with the same weight loadings.

## References

1. Sugihara, T.; Suzuki, M.; Komiya, M. M. U.S. Pat. 6,034,520 (2000).
2. Krueger, A. P. *Int J Biometeorol* 1985, 29, 205.
3. Suzuki, M.; Sugihara, T. U.S. Pat. 5,863,653 (1999).
4. Ge, X. L.; Li, E. CN Pat. 1,425,503 (2003).
5. Lee, W. S. KR Pat. 087,588 (2002).
6. Lee, W. S. KR Pat. 087,589 (2002).
7. Sasaki, K. J. U.S. Pat. 6,316,102 (2001).
8. Yuasa, I. M.; Hiramatsu, K. J. U.S. Pat. 5,981,063 (1999).
9. Ruan, D.; Zhang, L.; Zhang, Z.; Xia, X. *J Polym Sci Part B: Polym Phys* 2004, 42, 367.
10. Yeh, J. T.; Wei, W.; Hsiung, H. H.; Jiang, T. *J Polym Eng* 2006, 26, 117.
11. Yeh, J. T.; Wei, W.; Hsiung, H. H.; Jiang, T.; Chen, K. N. *J Appl Polym Sci* 2008, 109, 82.
12. Soo, S. M. U.S. Pat. 0,014,716 (2002).
13. Konishi, J. U.S. Pat. 6,365,192 (2002).
14. Shimoda, J. U.S. Pat. 5,906,638 (1999).
15. Yatagai, M.; Ito, R.; Ohira, T.; Oba, K. *Mokuzai Gakkaishi* 1995, 41, 425.
16. Hirata, Y.; Kyoto, Y.; Takase, H. U.S. Pat. 6,833,145 (2004).
17. Tatsumi, H. U.S. Pat. 6,509,294 (2003).
18. Konishi, J. E. U.S. Pat. 6,365,192 (2002).