ORIGINAL PAPER

Mechanical–Thermal Properties and VOC Emissions of Natural-Flour-Filled Biodegradable Polymer Hybrid Bio-Composites

Hee-Soo Kim · Byoung-Ho Lee · Hyun-Joong Kim · Han-Seung Yang

Abstract The mechanical–thermal properties and volatile organic compound (VOC) emissions of natural-flour-filled, biodegradable polymer bio-composites were investigated according to variation in porous inorganic filler types. At a porous inorganic filler content of 3%, the tensile and flexural strengths of the hybrid bio-composites were not significant changed. However, the coefficient of thermal expansion and thermal expansion of the bio-composites were slightly decreased. Furthermore, the incorporation of the porous inorganic materials into bio-composites slightly increased the E' values of the hybrid bio-composites over the entire temperature range, although the tan δ_{max} temperature (T_g) of the hybrid bio-composites was not significantly changed. At a porous inorganic filler content of 3%, the various odor and VOC emissions of the hybrid bio-composites were significantly decreased because the various oxidation and thermal degradation gases of the natural flour and matrix were absorbed in the pore structures of the porous inorganic fillers and thereby prevented the migration into the final products.

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Introduction

Recently, bio-composite materials reinforced with natural-fillers such as natural flour and natural fiber have been recognized as very attractive materials for application to biodegradable polymers in order to reduce the environmental problems associated with non-biodegradable plastic waste [1, 2]. These bio-composites, which are made using natural-fillers as reinforcing filler, have the advantages of easy availability, high specific strength and modulus, low density, low cost, low CO_2 emission, ease of surface modification, relative non-abrasiveness and biodegradability, compared to inorganic materials such as glass fiber, carbon fiber, synthetic fiber and talc, etc. [3, 4]. Bamboo flour (BF) is an abundant natural resource in Asia, and it can be renewed much more rapidly than wood flour (WF) due to bamboo's more rapid growth than wood. Because of this property, BF has gradually invaded wood forests and reduced the wood supply [5]. Synthetic non-biodegradable polymers such as polypropylene (PP), polyethylene (PE) and polystylene (PS) are widely used as matrix polymers in natural-filler-reinforced composite systems due to their relatively superior properties such as low density, easy processing and low cost [6]. However, these polymers are generally resistant to biological degradation due to their non-biodegradability and they therefore tend to accumulate in the natural environment. To solve this problem, recycling of these polymers is a better long-term solution than disposal, but only a minor portion of the plastics is recyclable and most still ends up in municipal burial sites [7, 8]. Therefore, the use of eco-friendly,

biodegradable polymers as a substitute for traditional, non-biodegradable polymers has received considerable attention in developing eco-friendly green composites and bio-composites [9].

Polylactic acid (PLA) and polybutylene succinate (PBS) are biodegradable, aliphatic thermoplastics which have recently been introduced commercially for products where biodegradability is wanted. PLA is a versatile polymer made from renewable agricultural raw materials, which are fermented to lactic acid. The lactic acid is then converted via a cyclic dilactone, lactide, ring opening polymerization to the wanted PLA [10]. PBS is chemically synthesized by the polycondensation of 1,4-butanediol and succinic acid with a melting point of about 110–120 °C (similar to LDPE), glass transition temperature (T_g) of about −45 to −10 °C (between PE and PP), tensile strength between PE and PP, and stiffness between LDPE and HDPE [11]. The main application areas of natural-filler-reinforced composites are automotive and building industries in which they are used in structural applications such as fencing, decking, outdoor furniture, window parts, roofline products and door panels, etc. [3, 12]. The present study investigated the use of porous inorganic fillers in natural-flour-filled, biodegradable polymer bio-composites in order to reduce the odor and volatile organic compound (VOC) emissions, and to improve the thermal properties. In recent years, with the increasing concerns about indoor air quality (IAQ), reducing the VOC emissions of the interior materials used as automotive parts and building has become widely recognized as an important factor for interior materials [13, 14]. However, little attention has recently been focused on the reducing odor and VOC emissions of natural-filler-filled, biodegradable polymer bio-composites. Furthermore, few studies have been reported on the use of porous inorganic filler as a reinforcing filler in the natural-flour-filled, biodegradable polymer, bio-composite hybrid system. In the melt mixing process of bio-filler-filled, thermoplastic polymer composites using a twin screw extruder, odor and various VOCs are emitted by the bio-filler due to the low degradation temperature of the natural-fillers [15]. In addition, VOC emissions are generated from the matrix polymer due to the random chain scission of the matrix polymer and oxidation resulting from the high manufacturing temperature [16, 17]. In the current work, we used porous inorganic fillers such as natural/synthetic zeolite, pozzolan and white clay to reduce the odor and VOC emissions and to increase the thermal properties of natural-flour-filled, biodegradable polymer bio-composites. These inorganic fillers, which are crystalline inorganic materials possessing a high SiO_2/Al_2O_3 ratio and a three-dimensional network system of large pore channels, have attracted significant technological interest [18]. These pores have variable openings, according to the structure type, which usually feature internal cavities of variable shape and diameter [19]. The aim of this study was to investigate the effect of porous inorganic filler type on the mechanical properties, thermal properties and VOC emissions of natural-flour-filled, biodegradable polymer, hybrid bio-composites. The investigation focused on reducing various odor and VOC emissions and the increasing thermal properties of the bio-composites by using porous inorganic fillers. The generated odor and VOC emissions of porous inorganic fillers-treated and non-treated bio-composites were measured using a gas chromatograph coupled to a mass selective detector (GC-MSD). In addition, the mechanical and thermal properties of porous inorganic filler-treated and non-treated bio-composites were investigated.

Experimental

Materials

Biodegradable Polymer and Natural Flour

PBS was prepared at Ire Chemical Ltd., South Korea, with an MFI of 25 g/10 min and a density of 1.26 g/cm³. PLA was supplied by Cargill-Dow Co. USA with an MFI of 15 g/10 min (190 °C/2,160 g) and a density of 1.22 g/cm³. The natural flours used as the reinforcing filler, BF and WF, were supplied by Hangyang Advanced Materials Co. South Korea. The particle size of BF and WF was 860–270, and 140 μm, respectively.

Porous Inorganic Fillers

Natural and synthetic zeolite, with an average particle size of over 7.7 and 9.7 μm, respectively, were obtained from G&C Bio Co., South Korea. Pozzolan in volcanic pozzolan ash form, with a particle size of over 70 μm, was obtained from AutoWin Co. White clay, with an average particle size of over 70 μm, was supplied by Donghae Chemical Co., South Korea. The chemical composition of the porous inorganic fillers was determined using an X-ray fluorescence spectrometer (Shimadzu XRF-1700, Japan).

Table 1 shows the chemical composition of the porous inorganic materials. These porous inorganic fillers have a high concentration of SiO_2 and Al_2O_3 elements, while synthetic zeolite has a high concentration of SiO_2 and MgO elements. Also, the BET surface area and average pore diameter were calculated by using a surface area analyzer (Micrometrics, model ASAP 2010). Table 2 shows the BET surface area and average pore diameter of the porous inorganic fillers.

Table 1 Content of porous inorganic materials

	Content of porous inorganic fillers
PBS: Natural Flour (BF, WF) = 70:30	3
PLA: Natural Flour (BF, WF) = 70:30	3

Compounding and Sample Preparation

BF and WF were oven dried at 105 °C for 24 h to adjust the moisture content to 1–3% and then stored in sealed polyethylene bags before compounding. PLA and PBS were blended with the BF, WF and porous inorganic fillers in a laboratory-sized, co-rotating, twin screw extruder using three general processes: melt blending, extrusion and pelletizing. The extruder barrel was divided into eight zones with the temperature in each zone being individually adjustable. Table 3 shows the content of porous inorganic fillers in the bio-composites. The temperature of the mixing zone in the barrel was maintained at 145 °C (PBS-based bio-composites) and 185 °C (PLA-based bio-composites) with a screw speed of 250 rpm. The extruded strand was cooled in a water bath and pelletized with a pelletizer. The extruded pellets was oven dried at 80 °C for 24-h and stored in sealed polyethylene bags to avoid unexpected moisture infiltration. The extruded pellets were injection molded into tensile (ASTM D 638) and three-point bend test bars (ASTM D 790) using an injection molding machine (Bau Technology, South Korea) at 145 °C (PBS-based bio-composites) and 185 °C (PLA-based bio-composites) with an injection pressure of 8 MPa and a device pressure of 10 MPa. After injection molding, the test bars were conditioned prior to testing at 50 ± 5% RH for at least 40 h according to the ASTM D 618-99.

Measurements

Mechanical Properties

The tensile test for the hybrid bio-composites was conducted according to ASTM D 638-99 with a Universal

Table 3 BET surface area and average pore diameter of the porous inorganic fillers

Sample	BET surface area (m^2/g)	Average pore diameter (nm)
Natural zeolite[a]	32.7	7.8
Synthetic zeolite[a]	2.1	21.9
Pozzolan	13.1	9.9
White clay	137.7	6.2

[a] Natural and synthetic zeolite, Ref [20]

Testing Machine (Zwick Co.) at a crosshead speed of 5 mm/min and a temperature of 24 ± 2 °C. The three-point bend test of bio-composites was carried out in accordance with ASTM D 790. The specimens had a span to depth ratio of 16:1 with a crosshead speed of 5 mm/min. Five measurements were conducted and averaged for the final result.

Thermal Properties

Dynamic Mechanical Analysis (DMA) The viscoelastic properties of the porous inorganic filler-treated and non-treated bio-composites were measured using a dynamic mechanical analyzer (DMA Q800, TA Instruments). Rectangular specimens having a size of 35.0 mm × 12.0 mm × 3.0 mm were used in the dual cantilever method. The measurements were performed at a frequency of 1 Hz and at a strain rate 0.1%. The temperature range was from −80 to 100 °C at a scanning rate of 2 °C/min. The storage modulus (E') and loss modulus (E'') and loss factor (tan δ) of the specimen were measured as a function of temperature.

Thermomechanical Analysis (TMA) The thermal expansion and coefficient of thermal expansion (CTE) tests of porous inorganic filler-treated and non-treated bio-composites were conducted using a thermomechanical analyzer (TMA 2940, TA Instruments) from ambient temperature to 120 °C at a heating rate of 2 °C/min in a high quality nitrogen atmosphere. Expansion mode with a constant compression load of 0.05 N was applied to the

Table 2 Chemical composition of porous inorganic materials

Element	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	MgO	CaO	Na_2O	K_2O	MnO	P_2O_5	LOI	Total
Natural zeolite[a]	59.9	16.2	0.4	3.9	1.9	4.4	2.9	1.6	0.1	0.1	9.4	100.8
Synthetic zeolite[a]	54.5	2.2	0.1	1.4	28.9	3.5	–	0.01	0.02	0.1	8.6	99.3
Pozzolan[b]	59.5	17.7	0.9	7.6	1.3	0.2	0.9	4.7	0.1	5.0	5.0	102.9
White clay	63.1	13.9	1.4	4.4	0.7	3.1	1.7	1.4	0.03	0.1	10.2	100.3

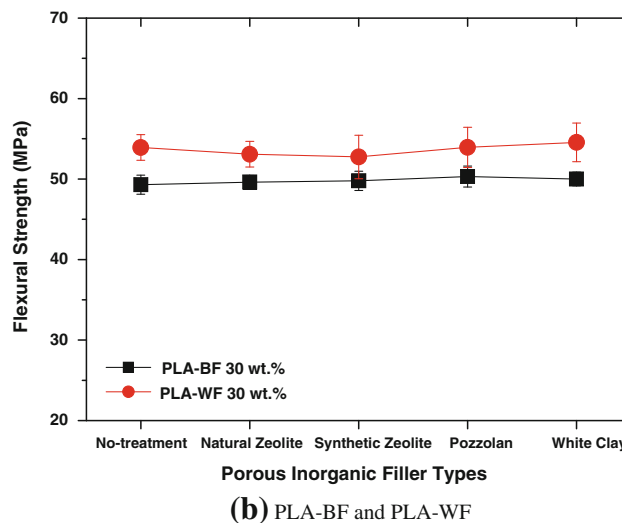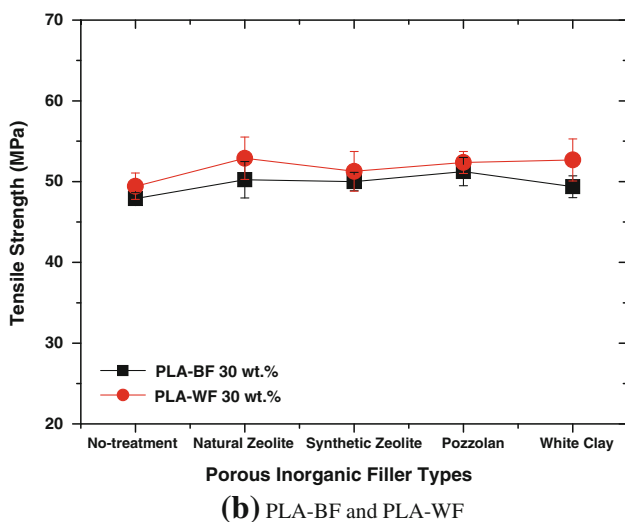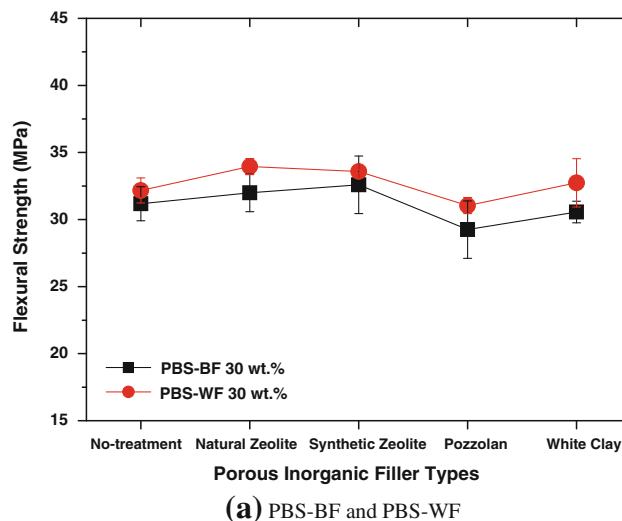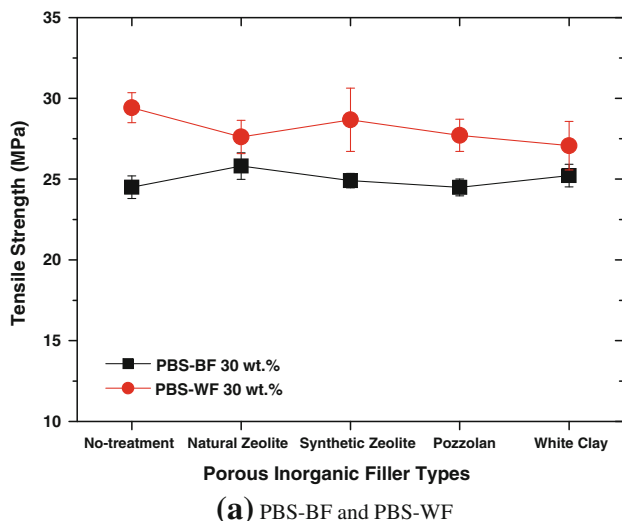[a] Natural and synthetic zeolite, Ref [20]

[b] Pozzolan, Ref [21]

Fig. 1 Tensile strength of BF- and WF-filled **a** PBS and **b** PLA hybrid bio-composites with different porous inorganic filler types

Fig. 2 Flexural strength of BF- and WF-filled **a** PBS and **b** PLA hybrid bio-composites with different porous inorganic filler types

specimen in the testing process. The specimens were cut into rectangular prisms of dimensions $5 \times 5 \times 3.2$ (mm).

VOC Analysis

GC-MSD Analysis The GC-MSD analysis of the porous inorganic filler-treated and non-treated bio-composites was performed on a CP-3800 gas chromatography/Saturn 2000 MSD with a CP SIL 5CB capillary column. The carrier gas was helium with a flow rate of about 1.2 ml/min. The GC oven temperature was initially held at 35 °C for 5 min, then raised to 250 °C at a rate of 8 °C/min and finally maintained at that temperature for 30 min. The bakeout time was increased to 30 min with a constant temperature of 200 °C. MS acquisition parameters included scanning from m/z = 30–650 amu in the electron impact (EI) mode automatically. The seven bio-composites were stored in

odorless polyester bags containing injected, high quality, nitrogen gas and maintained at room temperature for 24 h, after which the VOC emissions of the bio-composites was analyzed by GC-MSD.

Results and Discussions

Mechanical Properties

Figures 1 and 2 show the tensile and flexural strengths of BF- and WF-filled (a) PBS and (b) PLA hybrid bio-composites with different porous inorganic filler types. At a porous inorganic filler content of 3%, the tensile and flexural strengths of the hybrid bio-composites were largely unaffected, indicating that no interfacial interaction occurred between the hydrophilic inorganic porous filler

and the hydrophobic biodegradable matrix. In recent years, to enhance the interfacial adhesion between a hydrophilic filler and a hydrophobic biodegradable polymer, a coupling agent and surface modification of the hydrophilic filler have generally been used to modify the filler-matrix interface and thereby enhance the filler-matrix interface as a result of improving mechanical and thermal properties of bio-composites [11, 22, 23]. In addition, the tensile and flexural strength of the WF-filled PBS and PLA hybrid bio-composites was slightly higher than that of the BF-filled PBS and PLA hybrid bio-composites, due to the difference of the particle size of BF and WF. In a previous study, Kim et al. [24] reported that the tensile strength of WF-filled PP composites was higher than that of RHF-filled PP composites because WF showed a higher holocellulose and lignin content and better filler particle dispersion due to its smaller particle size. Thus, due to its smaller particle size, WF tends to form smaller agglomerates than the larger particle sized RHF. Therefore, this study concluded that the addition of porous organic fillers in the manufacturing process of bio-composites to reduce the odor and VOC emissions of the bio-composites did not negatively affect the mechanical properties.

Thermal Properties

Dynamic Mechanical Analysis (DMA)

DMA test methods have been widely used for investigating the structures and viscoelastic behavior of bio-composite materials. Figure 3 presents the variations of the E' of PBS-WF hybrid bio-composites with different porous inorganic filler types. With increasing temperature, the E' values of the bio-composites significantly decreased due to the softening and increased polymer chain mobility of the
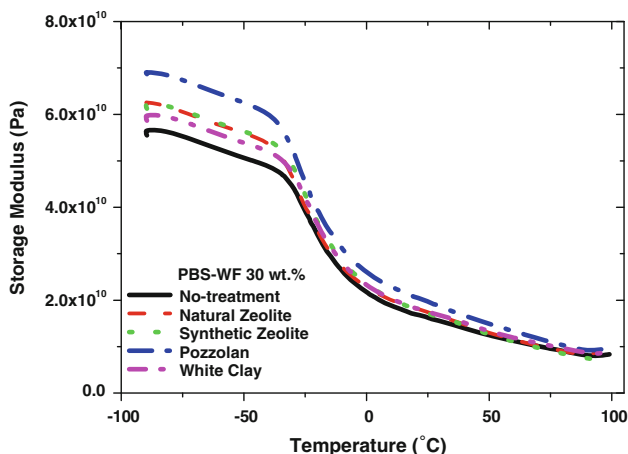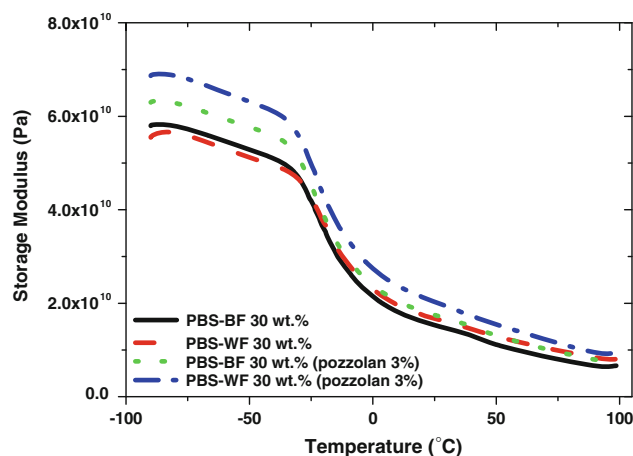
Fig. 4 Storage modulus of PBS-BF and PBS-WF hybrid bio-composites at a pozzolan content of 3%
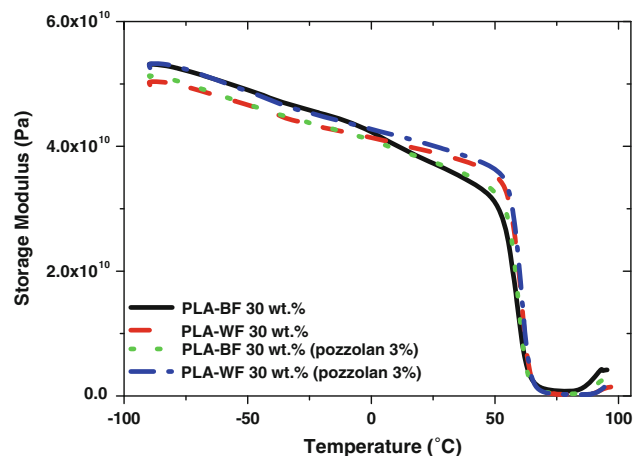
Fig. 5 Storage modulus of PLA-BF and PLA-WF hybrid bio-composites at a pozzolan content of 3%

matrix at higher temperatures. Also, the E' values of the PBS-WF hybrid bio-composites was slightly increased over the whole temperature range by the addition of porous inorganic materials into the PBS-WF bio-composites. This result was also seen in Fig. 4. This result was indicated by the ability of the porous inorganic fillers to produce a more rigid interface and the reinforcing effect imparted by the porous inorganic fillers which facilitated greater stress transfer at the interface in the PBS-WF bio-composites [25]. Also, the E' value of the pozzolan-treated PBS-WF bio-composites was the highest, which was attributed to greater particle size of pozzolan. The E' variations of the PLA-BF and PLA-WF hybrid bio-composites at the pozzolan content of 3% are shown in Fig. 5. The E' value of PLA-BF and PLA-WF bio-composites was significantly decreased about 60 °C, which was mainly related to the T_g of PLA. The main disadvantage of PLA is the ease of its deformation above T_g, as confirmed by the significantly decreased E' value of the PLA-BF and PLA-WF

Fig. 3 Storage modulus of PBS-WF hybrid bio-composites with different porous inorganic filler types

bio-composites. Also, the E' value of the pozzolan-treated PLA-BF and PLA-WF bio-composites was slightly increased due to the increasing stiffness of these bio-composites.

The temperature dependence of tan δ for the PBS-WF hybrid bi-composites with different porous inorganic material type is presented in Fig. 6. Tan δ is the ratio of E'' (viscous phase) to E' (elastic phase). With increasing temperature, the tan δ values of the PBS-WF hybrid bio-composites increased due to the increased polymer chain mobility of the matrix. The tan δ_{max} peak can also provide information on the T_g and energy dissipation of the bio-composite materials. The T_g is assigned to the energy dissipation possibilities across the free amorphous phase [6, 25]. Fig. 7 shows the tan δ_{max} temperature (T_g) of the
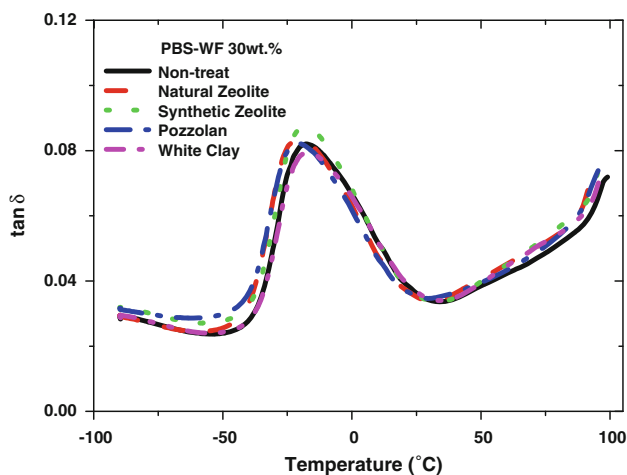

Fig. 8 TMA thermograms of the dimensional change of PBS-WF bio-composites as function of porous inorganic filler type

Fig. 6 Tan δ of PBS-WF hybrid bio-composites with different porous inorganic material types
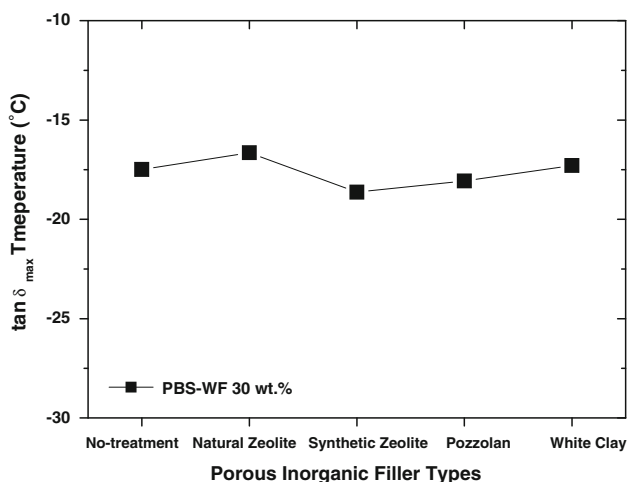

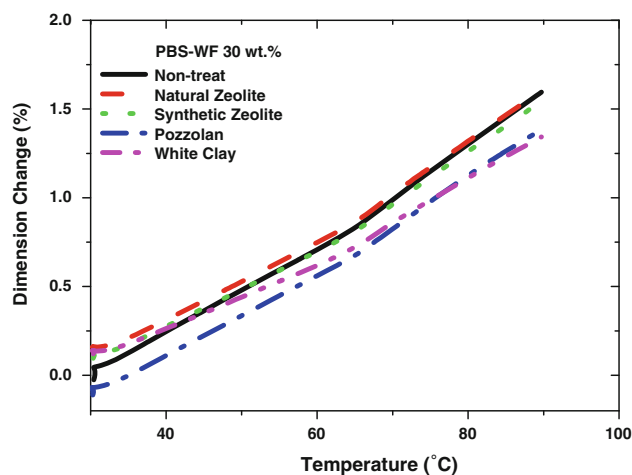Fig. 7 Tan δ_{max} temperature of PBS-WF hybrid bio-composites with different porous inorganic material type

PBS-WF hybrid bio-composites with different porous inorganic material types. The T_g of the PBS-WF hybrid bio-composites was not significantly changed by the addition of the porous inorganic filler. This result indicated that the porous inorganic materials were not affected by the T_g of the natural-flour-filled, bio-degradable polymer bio-composites.

Thermal Expansion Analysis

TMA expansion curves of the porous, inorganic filler-treated and non-treated PBS-WF bio-composites as a function of porous inorganic filler type are shown in Fig. 8, and the CTE values of the PBS-WF bio-composites are shown in Table 4. The TMA method for CTE measurement is useful for understanding the dimensional changes of bio-composite materials as well as the thermal stresses caused by increasing temperature [26, 27]. A lower CTE value of the bio-composites indicates that the bio-composites undergo lower dimensional change when exposed to cold or warm atmospheric change [26]. The thermal expansion and CTE value of the PBS-WF hybrid bio-composites was slightly decreased by the addition of 3 wt.% porous inorganic fillers, indicating that the incorporation of porous inorganic filler in the bio-composites is capable of preventing the thermal expansion of the bio-composites at high temperature due to the lower thermal expansion of the porous inorganic filler. In addition, the CTE value of the white clay-treated, PBS-WF bio-composite was the highest because the particle size of white clay was much higher than that of the other porous inorganic fillers. These results suggested that the addition of porous inorganic materials in the bio-composites is an effective method for reducing their thermal expansion and dimensional change.

Table 4 Comparison of the coefficients of thermal expansion (CTE) of PBS-WF bio-composites as function of porous inorganic filler type

Specimen	From 40 to 80 °C (μm/m °C)
PBS-WF 30 wt%	259.3
PBS-WF 30 wt% (Natural zeolite 3%)	244.9
PBS-WF 30 wt% (Synthetic zeolite 3%)	242.9
PBS-WF 30 wt% (Pozzolan 3%)	251.3
PBS-WF 30 wt% (White clay 3%)	212.4

VOC Analysis

GC-MSD Analysis

This study measured the odor and VOC emissions of natural-flour-filled, biodegradable polymer bio-composites at a porous inorganic filler content of 3%. Figure 9 shows the volatile odor compound emissions of porous inorganic filler-treated and non-treated, BF- and WF-filled, PBS and PLA bio-composites: (a) furfural emission, and (b) 5-methyl furfural emissions at the porous inorganic filler content of 3%. Odor production of the bio-composites can be caused by the matrix polymer or the natural-filler, or by a combination of the two. The matrix polymer can emit unwanted odors, caused by residual monomers or by decomposition products that occur in the production process of bio-composites due to the non-optimized heating temperatures [21]. Furfural and 5-methyl furfural are the main materials which generate the odor emitted by the bio-composites. Furfural can be produced from natural-filler materials rich in pentosan polymer from wood and natural-filler materials [28, 29]. BF and WF can emit odors caused by the thermal degradation of natural flour surfaces due to the low degradation temperature of BF and WF during the high temperature manufacturing process. The furfural and 5-methyl furfural emissions of porous inorganic filler-treated hybrid bio-composites were significant lower than those of non-treated bio-composites. This indicated that the volatile odor compounds and thermal degradation gases from the natural flour were absorbed at the pore structures of the porous inorganic fillers in melt mixing process of bio-composites using a twin screw extruder [17].

Tables 5 and 6 list the emission levels, as detected by GC-MSD, of various VOCs from porous inorganic filler-treated and non-treated BF-filled PBS and PLA bio-composites. The various VOC emissions of these bio-composites were mainly caused by the oxidation of the matrix polymer and the thermal degradation of natural flour during extrusion in the twin-screw extruder barrel. At the porous inorganic filler content of 3%, the various VOC emissions of the hybrid bio-composites significantly
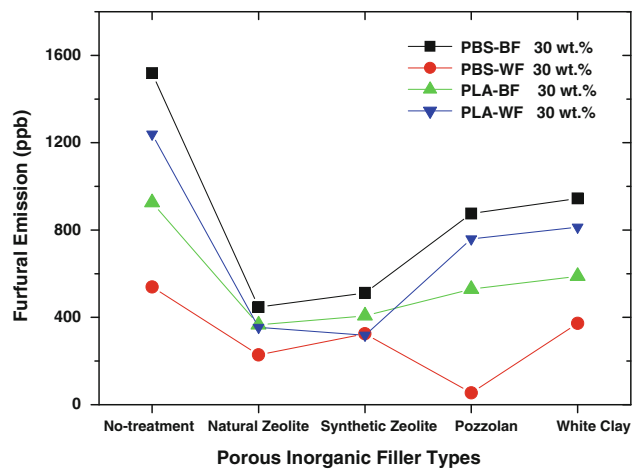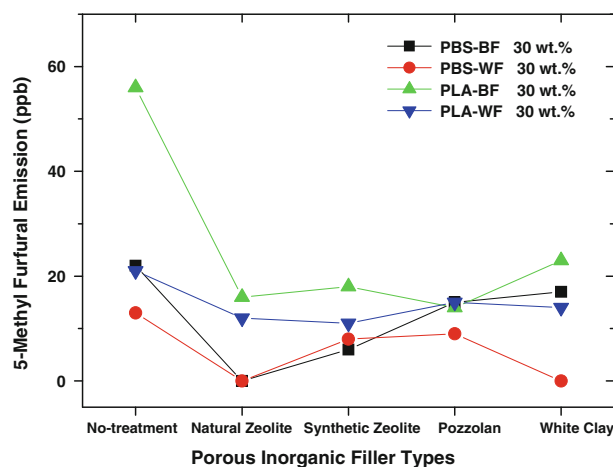

(a) Furfural emission

(b) 5-Methyl furfural emission

Fig. 9 Volatile odor compound emissions of porous inorganic filler-treated, BF- and WF-filled, PBS and PLA bio-composites: **a** furfural emission and **b** 5-methyl furfural emission

decreased due to the hybridization effect of the porous inorganic fillers, which was impossible in natural-flour-filled PBS bio-composites. The open porous structure of the porous inorganic fillers makes them active as adsorbents [16, 17]. This open porous structure could absorb the various VOC emissions and thermal degradation gases of the matrix and natural-flour that were generated during the manufacturing process of bio-composites.

The various VOC emissions of the natural zeolite- and pozzolan-treated hybrid bio-composites were lower than that of the synthetic zeolite- and white clay-treated hybrid composites. These results demonstrated the variation in pore size of the natural and synthetic zeolites, which is strictly connected with their structure [30]. Therefore, the present study results support the potential for the addition of porous inorganic filler as an effective method to reduce the odor and VOC emissions of natural-flour-filled,

Table 5 Various VOC emissions from porous inorganic filler-treated and non-treated, PBS-BF hybrid bio-composites

Detected item (VOC)	Sample				
	No-treatment	Natural zeolite	Synthetic zeolite	Pozzolan	White clay
Acetone	194	64	85	105	172
Dichloro methane	52	13	15	30	41
2-Butanal	16	0	2	8	7
Methyl ethyl ketone	30	20	22	17	24
2-Ethyl butanal	6	0	0	0	0
Ethyl acetate	33	9	8	15	23
Tetra hydro furan	15	11	8	12	10
3-Methyl butanal	11	4	4	7	3
Toluene	396	81	147	272	280
Hexanal	35	11	12	15	31
N,*N*-dimethylacetamide	520	400	324	423	185
m,p-xylene	74	14	24	45	60
Styrene	7	4	4	5	5
Benzaldehyde	76	38	28	42	40
1,2,4-TMB	59	34	41	50	37
1-Ethyl-4-methyl benzene	68	16	13	30	41
1,2,3-Trimethyl benzene	29	18	15	22	27
1-Ethyl-2-methyl benzene	276	154	167	159	208
1,3,5-Trimethyl benzene	164	95	70	72	117
2-Ethyl-1-hexanal	158	82	85	93	90
1-Ethyl-3-methylethyl benzene	81	45	42	40	25
1-Methyl propyl benzene	93	27	38	62	77

Table 6 Various VOC emissions from porous inorganic filler-treated and non-treated, PLA-BF hybrid bio-composites

Detected item (VOC)	Sample				
	Non-treat	Natural zeolite	Synthetic zeolite	Pozzolan	White clay
Acetone	166	87	119	141	162
2-Butanal	12	8	6	7	12
Methyl ethyl ketone	148	106	114	133	158
Tetra hydro furan	12	0	6	8	7
Acetic acid, 2-propenyl ester	99	45	37	62	66
3-Methyl butanal	65	19	21	26	42
Toluene	610	325	310	412	449
Hexanal	185	63	90	104	124
m,p-xylene	33	11	15	17	20
2-Heptanone	16	8	5	9	8
Heptanal	49	22	25	31	33
Pinene	12	10	11	8	14
2-Penyl furan	60	41	38	52	43
2-Ethyl-1-hexanal	49	36	27	48	37
1-Ethyl-3-methylethyl benzene	55	54	43	50	55
Limonene	11	7	5	9	9
Terpineol	27	27	26	12	12
Tridecane	47	25	33	32	42

biodegradable polymer bio-composites and thereby support their use as automotive interior and building interior materials.

Conclusions

At a porous inorganic filler content of 3%, the tensile and flexural strengths of the hybrid bio-composites showed very little change. The E' values of the hybrid bio-composites were slightly increased over the whole temperature range by the incorporation of the porous inorganic fillers but the tan δ_{max} temperature (T_g) of the hybrid bio-composites was not significantly changed. The thermal expansion and CTE value of the PBS-WF hybrid bio-composites was slightly decreased by the addition of 3 wt.% porous inorganic fillers. The various volatile odor compounds and VOC emissions of the hybrid bio-composites were caused by the presence of natural flour and matrix polymer in the manufacturing process of the bio-composites when run at non-optimized heating temperatures. At a porous inorganic filler content of 3%, the various odor and VOC emissions of the hybrid bio-composites were significantly decreased because the various oxidation and thermal degradation gases of the natural flour and matrix were absorbed into the pore structures of the porous inorganic fillers and thereby prevented from migrating into the final products. The porous structure of the inorganic materials was attributed to the ability of the pores to capture and absorb odor and VOC emissions and thereby reduce the odor and VOC emissions of the natural-flour-filled, biodegradable polymer bio-composites during the manufacturing process. Therefore, we suggested that the addition of porous inorganic fillers in the manufacturing process is an effective method to reduce the various odor and VOC emissions of natural-flour-filled, biodegradable polymer bio-composites.

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