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Soundproofing Properties of Polypropylene/Clay/Carbon Nanotube Nanocomposites

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ABSTRACT: In this article, polypropylene (PP)/clay/carbon nanotube (CNT) composites were prepared via a solution blending method. Sound transmission loss (STL), determined with an impedance tube, was used to characterize their soundproofing properties. The STL for the PP/4.8 wt % clay/0.5 wt % CNT composite was about 15–21 dB higher than that for pure PP at high frequencies (3200–6400 Hz) and about 8–14 dB higher at low frequencies (580–620 Hz). X-ray diffraction (XRD) and transmission electron microscopy (TEM) were used to study the crystallinity and the microstructure. A synergistic effect on the STL was established between the structure of the homogeneous dispersion and strong interfacial adhesion. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 130: 504–509, 2013

KEYWORDS: clay; properties and characterization; nanostructured polymers; molding; composites

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INTRODUCTION

Noise is a byproduct of technological development and is recognized as a component of industrial pollution. Noise comes from a variety of sources and has become a significant problem that can severely disrupt the activities of many people.[1–3] Consequently, research on noise reduction using sound insulating and absorbing materials has been carried out for a wide range of applications.

Conventional inorganic fillers, such as talc, mica, and glass fiber, are not well-suited for noise reduction applications because of their high densities, hardnesses, and ineffectiveness in thin sections. On the other hand, nanocomposites based on organic polymers and inorganic fillers have shown remarkable enhancements in soundproofing properties, as well as mechanical, thermal, and electrical properties, at low filler loadings.[4,5] Lee et al. investigated the soundproofing effect of nanoparticle-reinforced polymer composites such as acrylonitrile butadiene styrene (ABS)/carbon nanotube (CNT) composites.[6] They reported that the sound transmission loss (STL) at 3400 Hz of ABS/CNTs at 15 vol % CNTs was 21.7% (4.1 dB) higher than for pure ABS. Moreover, Lee et al. studied the mechanical properties and the sound insulation effect of ABS/carbon black composites.[7] They reported that the STL of these composites was more than 10% higher than that of pure ABS. Polymer/nanoparticle composites have become a favorite topic for academic and industrial research because of their greatly improved properties and relative ease of preparation. The enhanced properties of such nanocomposites have been attributed to synergistic effects between the polymer matrix and the nanofiller.[8]

Polypropylene (PP) is one of the fastest growing thermoplastic materials because of its low price, low density, and ease of processing.[9–11] PP/CNT composites have been extensively studied because of their improved mechanical, electrical, and other properties.[12,13] PP/clay nanocomposites have potential use in the automotive sector.[14–16] Yan et al. investigated the soundproofing behavior of nanoclay-reinforced PP composites.[17] They verified that the sound insulation efficiency of these composites increased with increasing loading of nanoclay up to 7 wt %.

In this study, the PP matrix was reinforced with clay and CNT fillers to obtain better soundproofing properties than reported previously. The specimens were prepared by solution blending, and their soundproofing properties were measured using an impedance tube.[18,19] In addition, the microstructures of PP/clay/CNT composites were evaluated by X-ray diffraction (XRD) and transmission electron microscopy (TEM) to establish a micro-mechanism for the improvement in the soundproofing.

EXPERIMENTAL

Materials

Type HJ400 PP with a density of 0.91 g/cm3 from the Samsung Co. was selected as the polymer matrix material in this study.

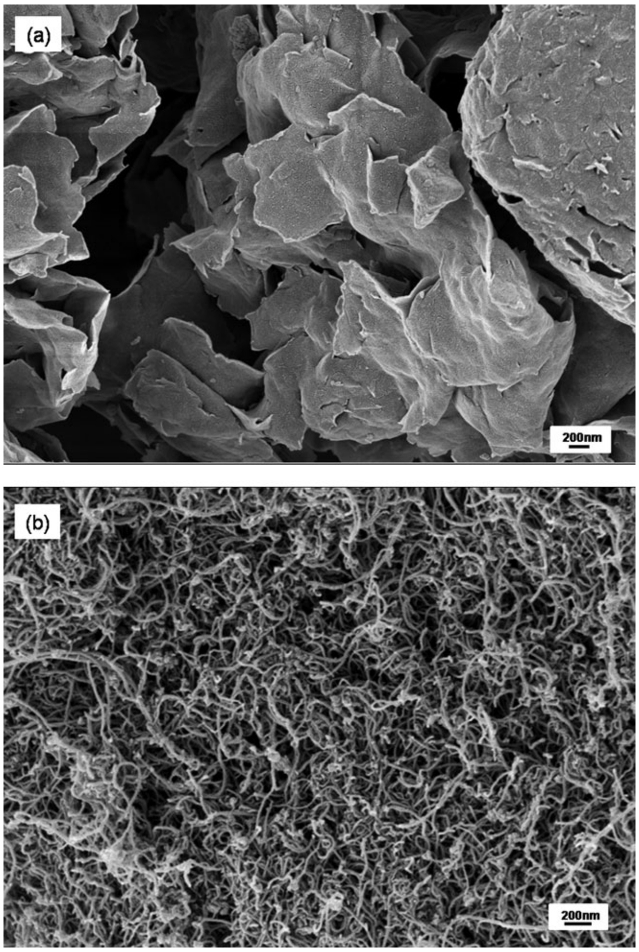

Figure 1. FE-SEM images of (a) clay and (b) CNTs particles.

Two kinds of fillers were used as reinforcing materials. Cloisite 15A clay with a density of 1.66 g/cm^3 and a particle size of 13 μm was obtained from Southern Clay Products. Type MWCNT CM-95 CNTs with a density of 1.8 g/cm^3 and a diameter of 15 nm were obtained from Hanwha Nanotech Co. Xylene and ethanol were purchased from Daejung Chemical & Metals. Field-emission scanning electron microscopy (FE-SEM) images revealing the morphologies of the clay and CNTs are shown in Figure 1(a,b).

Preparation of Composites

The nanocomposite powders were prepared by a solution blending method, as shown in Figure 2. First, PP was dissolved in xylene, and then the CNTs were poured into the solutions in xylene solvent at 0.1, 0.5, and 0.7 wt %, after that 0.9, 4.8, and 6.5 wt % clay were added. The blending was done at 135°C with magnetic stirring at 300 rpm for 2 h after which time the mixture was filtered. The retained precipitate was dried in an oven at 110°C for 4 h to evaporate the solvent. The powders were finally molded by heating in a press at 230°C and 35 MPa.

Characterization

STL was used to gauge the soundproofing properties of a composite. STL is defined as the difference between the sound power levels of the incident sound and the transmitted sound.[20] STL intensity is described by decibels (dB) according to eq. (1), where I_i is the incident acoustic power and I_t is the transmitted acoustic power[21]:

$$\mathrm{STL(dB)} = 10\log\frac{I_i}{I_t} \tag{1}$$

The STL values were measured at room temperature by the impedance tube method, as shown in Figure 3. The test set-up consisted of four 0.25-inch Brüel & Kjær (B&K) 4196 microphones, a B&K Type 4206 impedance tube, a B&K Type 2690 Nexus conditioning amplifier, an HP 35670A frequency analyzer, and LabView Version 7.0 software.[22] Two types of tubes were used: a 29-mm-diameter tube (small tube) for high frequencies (500–6400 Hz) and a 100-mm-diameter tube (large tube) for low frequencies (100–1600 Hz). The results are reported as the average of three samples with error bars.

STLs were measured for composites containing different fillers at various filler loadings. Composites of PP, which are commonly used in automotive and other applications, were used as benchmarks and compared with various PP/CNT, PP/clay, and PP/clay/CNT composites. The filler concentrations for each composite were 0.9, 4.8, and 6.5 wt % clay, 0.1, 0.5, and 0.7 wt % CNTs.

XRD measurements were performed with a Bruker AXS D8 Advance diffractometer with Cu Kα radiation ($\lambda = 0.1504$ nm) at room temperature. The instrument was operated at 40 kV and 40 mA. The XRD patterns were evaluated from 10° to 50° (2Θ) at a scanning rate of 2°/min.

TEM was used to evaluate the dispersion of the clay/CNT hybrid fillers in the PP matrix. A Zeiss EM 912 Omega instrument was used, operating at an accelerating voltage of 120 kV. A thin layer, about 70 nm thick, was sectioned from the crystallized sample at −100.0°C using an ultramicrotome equipped with a diamond knife.

RESULTS AND DISCUSSION

Soundproofing Properties

Using soundproofing materials is one of the simplest techniques for noise reduction. STL (reported in dB) is the usual measure of the soundproofing property of a material. A larger STL indicates a better soundproofing material.[23]

Many researchers have reported that PP/CNT composites have remarkable mechanical and thermal properties even at very low CNT loadings, e.g., under 1 wt %. There are many promising applications in automobiles, aerospace, packaging, and electronics for nanocomposites based on CNTs.[12,24–27] This work focuses on the sound barrier properties of PP/clay/CNT composites.

Figure 4 depicts the soundproofing properties of the PP/CNT composites as a function of the CNT content for high frequencies (29-mm-diameter specimens) and low frequencies (100-mm-diameter specimens). The results show that all samples containing CNTs were much better sound barriers than pure PP over all frequencies tested. In particular, the highest

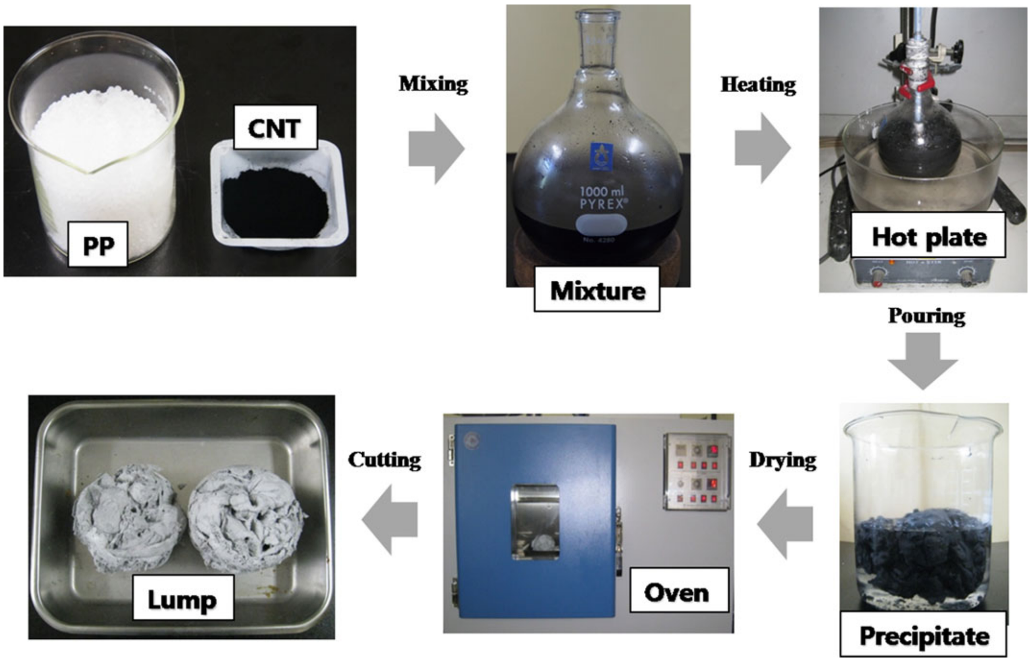

Figure 2. Manufacturing process of PP/filler composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

STL value was measured for the PP/0.7 wt % CNT composite. Thus, very low loadings of CNT particles in the PP matrix can dramatically improve the soundproofing properties.

PP/clay composites have the laminar structure and high aspect ratio, resulting in a large contact area available for interaction with the PP matrix. This results in composites exhibiting excellent mechanical properties, heat resistance, and dimensional stability.[9,10,14,28]

Figure 5 shows the soundproofing properties of the PP/clay composites as a function of the clay content. The PP/clay composites consistently yielded STL values higher than those of the pure PP matrix over the entire frequency range. Moreover, STL increased with increasing clay loading. The highest STL was measured with the PP/6.5 wt % clay composite.

A synergy was observed between the clay and the CNTs when combining 3–5 wt % clay with 0.4–1 wt % CNTs.[29,30]

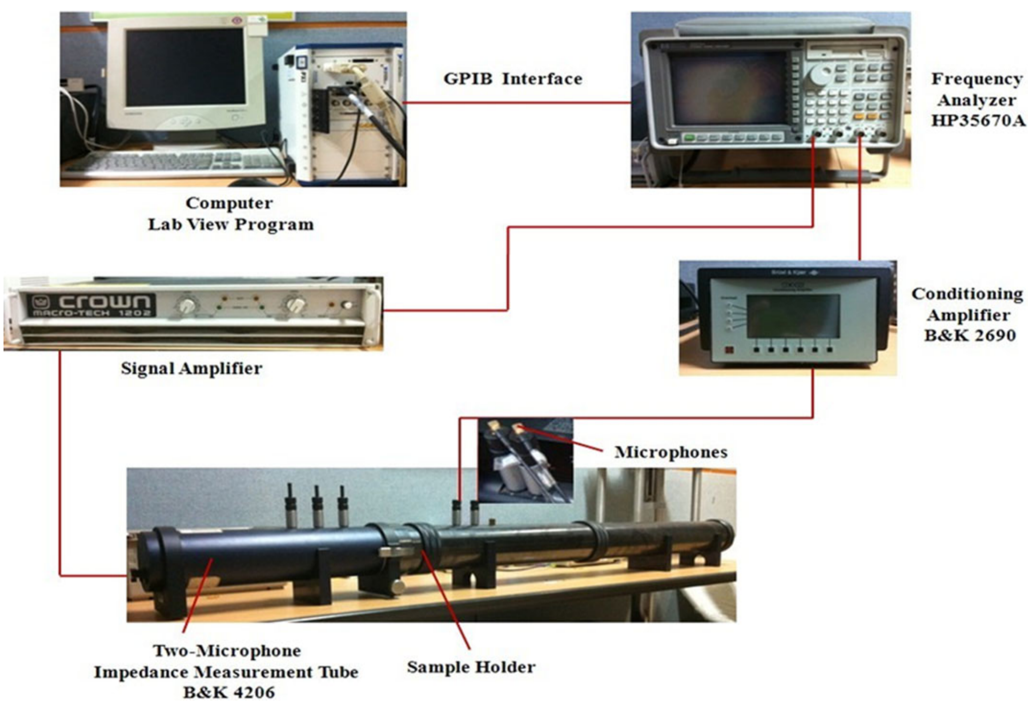

Figure 3. Impedance tube set-up for the STL measurements. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

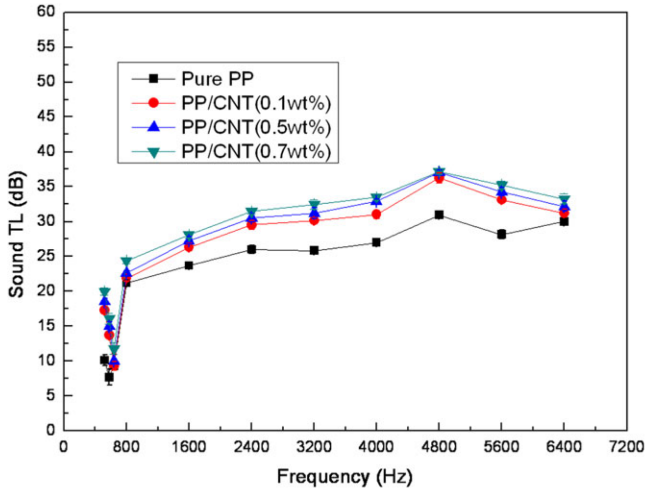

Figure 4. STL of PP and PP/CNT composites (520–6400 Hz). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

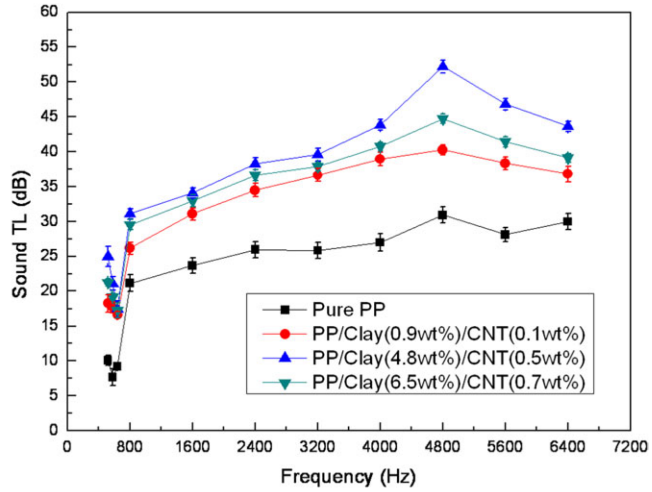

Figure 6. STL of PP and PP/clay/CNT composites (520–6400 Hz). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The soundproofing properties of these PP/clay/CNT composites as a function of filler content are shown in Figure 6. The STL gradually increased with increasing filler loading. However, in this test, the PP/4.8 wt % clay/0.5 wt % CNTs composition had enhanced soundproofing compared with the PP/6.5 wt % clay/0.7 wt % CNTs composite over the frequencies tested.

The soundproofing properties of selected PP/CNT, PP/clay, and PP/clay/CNT composites were compared with pure PP as function of filler content (Figures 3–6). The STL of pure PP, PP/0.7 wt % CNTs, PP/6.5 wt % clay, and PP/4.8 wt % clay/0.5 wt % CNT composites are compared in Figure 7. All specimens performed better than the PP matrix. The PP/4.8 wt % clay/0.5 wt % CNTs composite is notable because it had the STL values of 15–21 dB at 3200–6400 Hz and 8–14 dB at 580–620 Hz higher than those of pure PP. Automotive parts made from PP/clay/CNT composites would be about 25% lighter than conventional materials such as PP/talc, which are 10–20 wt % optimum loadings,[31,32] and would therefore help to improve fuel efficiency, even small amounts of clays or CNTs.[4,5] Thus, it is important to identify the reason why PP/clay/CNT composites have the best soundproofing properties.

Microstructure Analysis

Good dispersion of the nanofiller and strong interfacial adhesion between the nanofiller and the matrix are key requirements to achieve high-performing nanocomposites.[29] The PP crystal structure and the dispersion state of the clay and CNTs in the PP matrix were therefore measured using XRD and TEM.

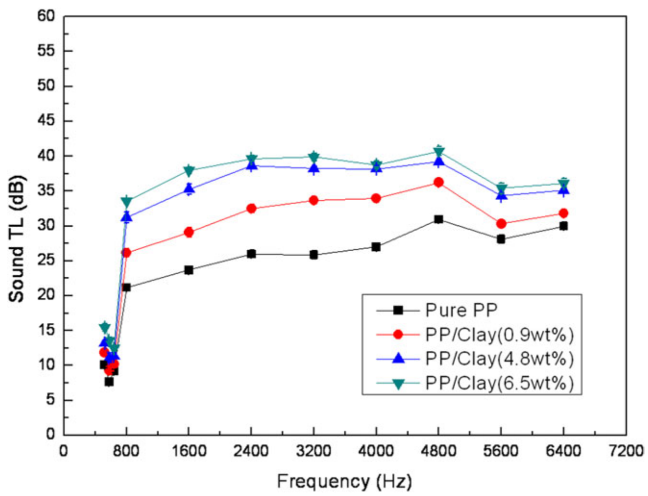

Figure 5. STL of PP and PP/clay composites (520–6400 Hz). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

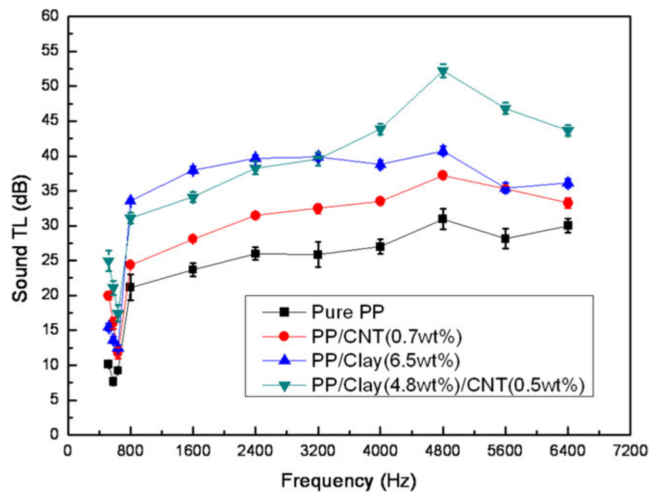

Figure 7. Comparison of the STL values of pure PP, PP/CNT, PP/clay, and PP/clay/CNT composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

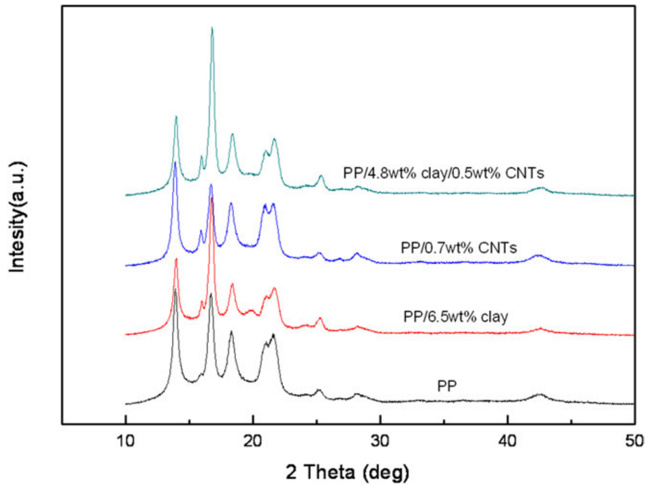

Figure 8. XRD profiles of PP, PP/clay, PP/CNT, and PP/clay/CNT composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

XRD was used to investigate the change in the crystal structure of the PP as a result of the added clay and CNTs. Figure 8 shows the XRD patterns of PP, PP/CNTs, PP/clay, and PP/clay/CNTs for different loadings of the fillers. Four peaks are shown in the XRD profiles of pure PP that correspond to the α-form. The addition of only 0.5 wt % CNTs and the presence of clay did not change the crystallinity.[12,33–35] This clearly indicates that small amounts of clay and CNT fillers do not affect the crystal form of the PP in the PP/CNT and PP/clay/CNT nanocomposites. Any change in soundproofing should thus be governed by the amounts of CNTs and clay inside the matrix and their dispersion levels.

The dispersion quality of the CNTs in the PP matrix is one of the most important factors influencing the mechanical and electrical properties of the composites. The addition of clay fillers improved the dispersion of the CNTs.[36] TEM was used to evaluate the dispersion state, the nanostructure features, and the synergistic reinforcing mechanism of clay and CNTs in the PP matrix for the soundproofing effect.

TEM images revealing the morphology and structure of the PP/0.7 wt % CNT, PP/6.5 wt % clay and PP/4.8 wt % clay/0.5 wt % CNT composites are shown in Figure 9. Figure 9(a) shows a TEM micrograph of a PP/CNT composite. One-dimensional CNTs are indicated by black arrows. CNT fillers are well dispersed into the PP matrix. CNTs have high mechanical strength, low specific gravity, and high aspect ratio.[12,27,37] Figure 9(b) displays the dispersion state of the clay in the matrix. The clay particles were dispersed homogeneously.[4,38] Good dispersion of the clay particles affects the synergies between properties, including the soundproofing effect.[39] Figure 9(c) illustrates the distribution of the clay and the CNTs in the PP matrix. There are no large clusters of CNTs and clay in the matrix. CNTs are randomly dispersed and connected to the clay layers. These images indicate that the PP/clay/CNT composites were successfully prepared through the solution blending method.[36,40] The PP chains are entangled and wrapped around the two-dimensional structure of the clay platelets and the one-dimensional structure of the CNTs. The dark areas represent the clay/CNT hybrid fillers, and the light areas represent the PP matrix. This three-way interaction improves the conversion of sound energy into heat energy and thereby improves the STL. Thus, the synergistic effect between the homogeneous dispersion and strong adhesion of the CNTs and clay platelets enhances the soundproofing performance of the composites.[29,41,42]

CONCLUSIONS

Nanocomposites were successfully prepared using a solution blending method. This technique provided a homogeneous dispersion and good interfacial adhesion of clay and CNTs in a PP matrix. STL was used to gauge the soundproofing ability of the various PP/clay/CNT composites. The PP/4.8 wt % clay/0.5 wt % CNT composite had the best soundproofing property; the STL exceeded that for pure PP by about 15–21 dB over 3200–6400 Hz and by about 8–14 dB over 580–620 Hz. These results indicate that the clay and CNT hybrid nanofillers remarkably enhance soundproofing properties. In particular, these PP/clay/CNT composites exhibited better soundproofing than composites reported in previous studies[6,17] that contained only one type of nanofiller, such as CNTs and clay. The soundproofing property depends strongly on the homogeneous dispersion of clay and CNTs in the PP matrix. The structures of PP and filler dispersion were investigated using XRD and TEM.

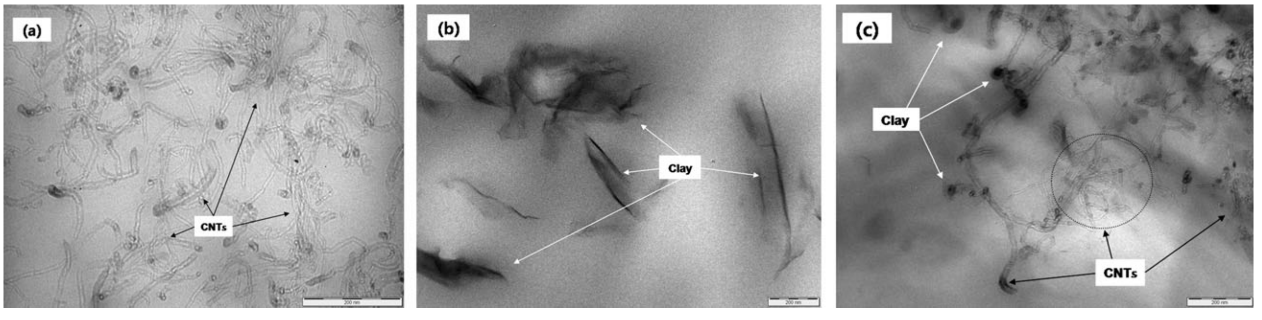

Figure 9. TEM micrographs of (a) PP/CNT, (b) PP/clay, and (c) PP/clay/CNT composites.

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