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공학박사 학위논문

Soundproofing and Mechanical Properties of Nanofiller-reinforced Polypropylene Composites

나노 필러로 강화된 폴리프로필렌 복합재의 흡음 및 기계적 특성

2015년 8월

서울대학교 대학원
기계항공공학부
양 준

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2015 년 8 월

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Abstract

Soundproofing and Mechanical Properties of Nanofiller-reinforced Polypropylene Composites

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Noise generally refers to unwanted sound or sound pollution which can cause serious harm to the human body and society. To prevent noise with sound absorbing materials has attracted much attention in many engineering fields nowadays. Recently, polymer absorbing materials have been fabricated for soundproofing application in many aspects of daily life. The advantages of such polymer absorbing materials are their high performance on soundproofing effect and internal properties such as light weight, specific strength, and low cost.

A kind of nanocomposite with clay reinforced polypropylene (PP) has been developed in order to investigate and understand its soundproofing and mechanical property. Clay is used as the reinforcement filler due to its nanometer dimension layered silicate which can dramatically change the mechanical and other performances at even very low filler concentration after homogeneous dispersion inside the host matrix. PP is used as the matrix material due to its attractive combination of low density and high heat distortion temperature.

The specimens made of this PP/Clay composites by injection molding machine are tested for their soundproofing property through impedance tube for sound transmission loss (TL) measurement. An optimum concentration of clay for proper performance ratio inside PP matrix is investigated in detail. Soundproofing property of pure PP, PP/Clay(0.9wt%), PP/Clay(4.8wt%), PP/Clay(6.5wt%) composites is discussed as a function of clay content with 29*mm* diameter specimens for high sound frequency. The soundproofing efficiency increases with clay content increasing from 0 to 6.5wt%. Compared with pure PP, average 8~20*dB* sound TL increases for PP/Clay(6.5wt%) composites at 3200~6400*Hz*. In general, 6.5wt% clay is the optimal amount for reinforcement to PP and this PP/Clay(6.5wt%) composite shows the best soundproofing. For mechanical properties, more clay filler specimens have higher storage modulus and loss modulus which can show better plastic property.

A mathematical equation for expressing the affecting factors of soundproofing property is developed with variables of sound frequency and clay weight percentage. Moreover, this equation shows perfect fitting curves which are similar to the results from experiment data. In addition, this kind of PP/Clay composites can be used for real products fabrication such as relay package and automotive interior parts.

Keywords: PP/Clay, Soundproofing, Mechanical property, Synergistic effect, Equation

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Chapter 1 Introduction

1.1 Overview

Noise generally means sound pollution which can cause serious harm to human body and society. It disturbs people and makes it difficult to hear wanted sounds. Acoustic noise can be anything from quiet but annoying to loud and harmful. To prevent noise with sound absorbing materials is one of the best ways for products fabrication in engineering field and has attracted much attention from many researchers. It means when sound transfers in such sound absorbing materials, energy loss and reflected energy must be maximized to minimize the transmitted energy. Therefore, the maximum reflection of incident sound leads to the increase of sound absorbing ability [1-5]. Among these sound absorbing materials, there are lots of attempts to reduce noise pollution by using polymer based composites such as PVC and polypyrrole nanometer additive [6-7]. These composites are composed with matrix of polymer and filler additive with particle size nanometer.

Recently, composites of nanofillers in polymer matrices have been widely studied for a variety of applications. Even small amounts of nanofillers, when mixed into an appropriate polymer matrix, can improve the stiffness, dimensional stability, mechanical properties, and sound-insulating ability of the neat polymer.

Currently, polymer absorbing materials have been fabricated for soundproofing application in various aspects of daily life. The advantages of such polymer absorbing materials are their high performance on sound absorbing effect with light weight, specific strength, and low cost at the same time. Anyway, polymer/nanofiller composites have become a welcome research topic in nowadays because of their remarkable improved properties and relatively easy preparation procedures [8-10].

In automotive applications, soundproofing materials are placed in the dashboard and center fascia of a vehicle's interior to minimize the influx of noise pollutants from the engine while simultaneously reducing electromagnetic interference to prevent malfunctioning of delicate safety equipment [11].

In this study, polymer composites with different nanofillers were synthesized and evaluated for their soundproofing properties by sound transmission loss (TL) measurement through impedance tube. In addition, the microstructure of these nanofiller-reinforced PP composites are observed and analyzed by TEM images in order to understand the relationship between soundproofing property and mechanism of the specimens. Moreover, a mathematical equation was finally built for expressing what kind of relative parameters and how they affected soundproofing property mutually.

1.2 Nanocomposites

Nanocomposites are a relatively new class of materials incorporating particles of various shapes and sizes. Typical particle dimensions are a few nanometers (nm). Nanocomposites composed of a traditional polymer matrix with various nanofillers boast enhanced mechanical, electrical, and thermal properties [12-17].

Nanocomposites can be classified into (A) 0-dimensional filler (atomic clusters, e.g. Silica), (B) 1-dimensional filler (e.g. CNT), (C) 2-dimensional filler (nanoscale layered structure, e.g. Clay platelet), and (D,E) 3-dimensional filler (filler networks and hybrid filler) nanostructures, as shown in Figure1.1.

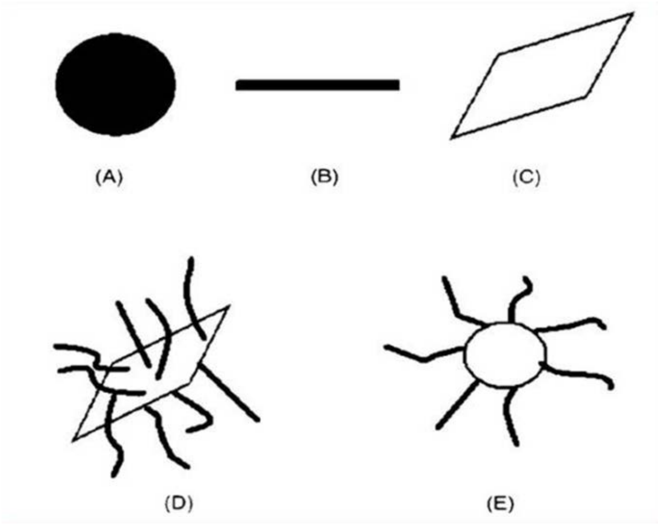


Figure1.1 Nanocomposites classification[18]

Nowadays, traditional filler materials such as talc, glass fiber, and wood flour are gradually being replaced by nanofillers such as nanoclay, CNT, and carbon black (CB). And proper selection of nanofiller with right amount to reinforce polymer matrix materials can bring much advantage to these kinds of nanocomposites with better performance and enhance on electrical, thermal, acoustic, and mechanical properties.

1.3 Soundproofing properties and materials

The acoustic energy of a material can be separated into the incident sound energy, reflected sound energy, absorbed sound energy, and transmitted sound energy which is shown in Figure1.2.

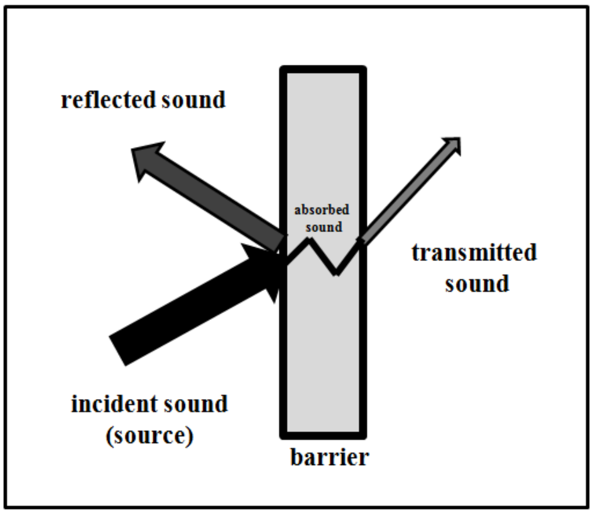


Figure1.2 Diagram of total acoustic energy[19]

Sound insulation is defined as the amount of acoustic energy incident reflected or absorbed rather than transmitted. The absorbed or reflected sound energy should be maximized while minimizing the transmitted sound energy should be minimized relatively for a good acoustic insulator. As energy loss through absorption and reflection is limited due to the thickness of the material, the most efficient way for increasing soundproofing performance of a material is to reflect the incident sound energy toward the direction of incidence. The soundproofing property is measured by sound TL which is defined as the difference between the sound power level of the incident wave and the transmitted sound intensity [19]. In mathematics, it is defined as the logarithmic ratio of the incident acoustic power (I_i) to transmitted acoustic power (I_t) as shown in Equation (1). Sound TL of the specimens is measured by two serially located microphones attached to an impedance tube, as shown in Figure 1.3 [20-22].

$$TL(dB) = 10\log\frac{I_i}{I_t} \quad (1)$$

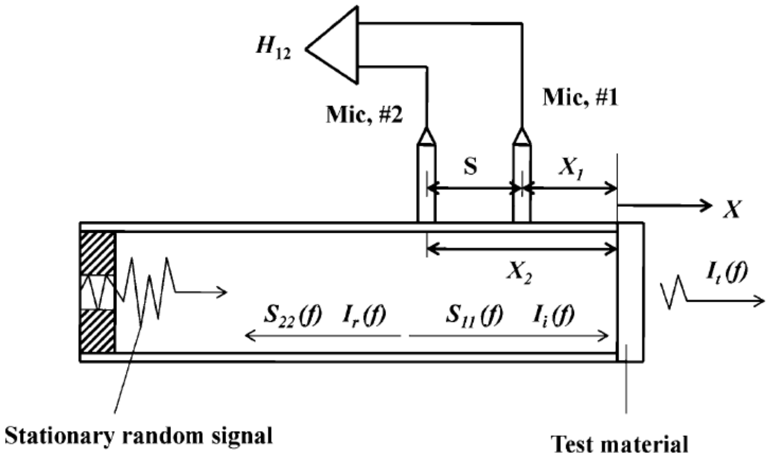


Figure1.3 Schematic of impedance tube [23]

Numerous studies across a variety of fields have explored soundproofing performance based on the above theories. For example, soundproofing materials are used in automobiles to minimize the influx of sound pollution into the vehicle interior. A variety of materials have being studied to enhance noise reduction and improve the fuel efficiency of lightweight materials for use in the automotive sector [24, 25].

Soundproofing materials have good ability for preventing sound transmission through sound absorption or insulation. Nowadays, lots of soundproofing materials are widely used in various engineering fields as shown in Figure 1.4.

Figure1.4 Soundproofing materials

1.4 Mechanical properties

Mechanical properties are very important for new materials which will be made into products for real application. Elementary mechanical properties include density, Young's modulus, elongation, and Poisson's ratio which reflect inherent characteristic such as elasticity, plasticity, strength, hardness, and ductility of this kind of new composite materials. Adding nanofiller particles into matrix polymer materials can change inner crystal structure of matrix polymer and alter its mechanical properties [26-30].

Chapter 2 Materials and specimens fabrication

2.1 Overview

There are lots of attempts to reduce noise pollution by using polymer based composites such as PVC and polypyrrole nanometer additive. Recently, polymer insulation and absorbing materials have been fabricated for soundproofing application in many aspects of daily life. The advantages of such polymer insulation and absorbing materials are their high performance on sound insulation effect and internal material properties such as light weight, specific strength, and low cost. In this study, PP matrix was reinforced with talc, clay, and CNT nanofillers to evaluate the soundproofing and mechanical property for use in automotive components.

2.2 Polypropylene

A polymer is a large molecule or macromolecule, composed of many repeated subunits. Because of their broad range of properties, both synthetic and natural polymers play an essential and ubiquitous role in everyday life. Due to macromolecule and dispersibility of molecule distribution, polymers have various high abilities on acoustic, thermal, electrical and mechanical properties. PP is one of commercial thermoplastics with attractive combination of low density and high heat distortion temperature. It has widespread availability on packaging, labeling, textiles, stationery, containers, and automotive components due to high melting point, hardness, and high polymerizability. And recently, many nano-sized fillers have been used to enhance the mechanical, electrical, acoustic, and thermal properties of PP [31-36]. Figure 2.1 shows the molecular structure and original shape of PP used in this research.

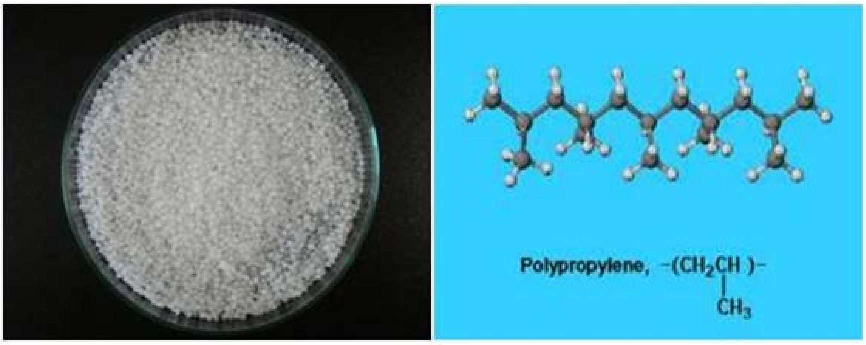


Figure 2.1 Molecular structure of pure PP

2.3 Fillers

Nanofillers are used to improve the performance of a polymer matrix with only a small amount which could be actively investigated for reducing the weight and enhancing the mechanical property of these composite materials. Recent studies are focusing on the research of soundproofing property of polymers reinforced by appropriate nanofillers. And many researchers are interested in improving the soundproofing property by incorporation of an appropriate nanofiller [22,37,38]. In this research, many kinds of nanofillers were studied for their soundproofing property and the results comparison were shown in Table 2.1 with 100*mm* diameter specimen at 520~640 *Hz*. As talc, clay and CNT reinforced PP matrix can produce more sound TL among these nanofillers, research on soundproofing effect of PP matrix material with nanofillers such as talc, clay, and CNT was investigated specifically in this research.

Table 2.1 Sound TL of different nanofiller reinforced PP composites

Nanofiller	Type	Production	Sound TL
Graphite(0.7wt%)	V-cond20	GK Ltd, Germany	1.1~1.7 *dB*
White crystal(4.8wt%)	Rock crystal	Sejin Company, Brazil	2~3 *dB*
Tourmaline(6.5wt%)	Cyclosilicate	Sejin Company, Kazakhstan	1.5~2.5 *dB*
Charcoal(0.9wt%)	Bamboo charcoal	Myanmar	0.3~1 *dB*
Talc(15wt%)	KR-2000	Kyoungki Chemical	3~5 *dB*
Nanoclay(6.5wt%)	Cloisite 15A	Southern Clay Pruducts	3.5~5 *dB*
CNT(0.7wt%)	MWNT CM-95	Hanwha Nanotech	5~10 *dB*

2.3.1 Talc

Talc is a mineral composed of hydrated magnesium silicate which has layered structure as shown in Figure 2.2. A composite of PP and talc is stronger and stiffer than pure PP [39-41].

Talc is used in many industries such as paper making, plastic, paint and coatings, ceramics, etc. However, due to large proportions of talc, this kind of PP/talc composites are relatively heavy than other nanofiller-reinforced PP composites [42]. Figure 2.3 shows the shape and molecular structure of talc used in this research.

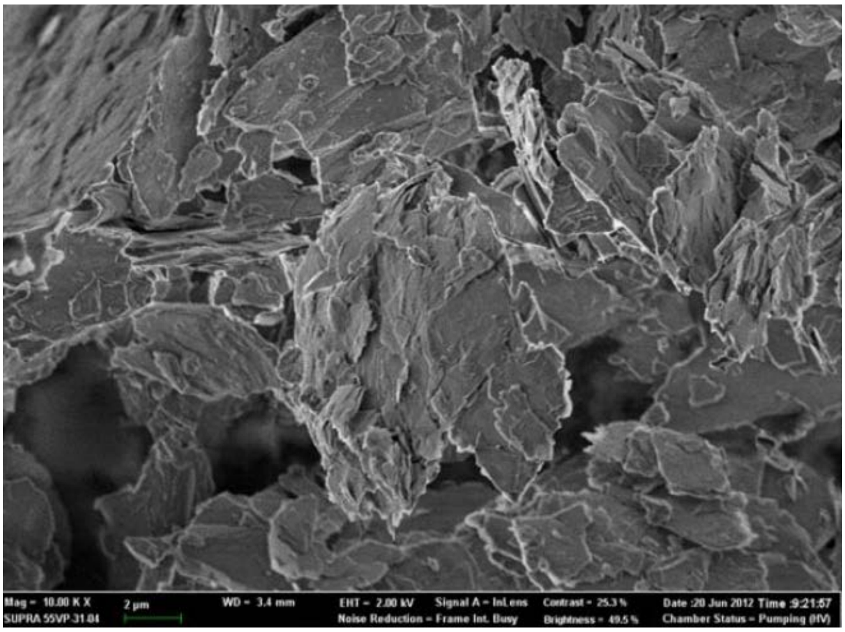


Figure 2.2 FE-SEM image of talc

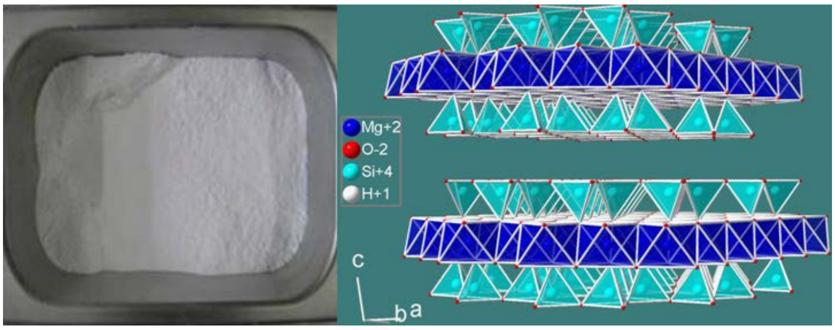


Figure 2.3 Talc and its molecular structure [40]

2.3.2 Nanoclays

Nanoclay typically exhibits a layered structure with 1-nm layer thickness, as shown in Figure 2.4. Nanoclay is a naturally occurring material composed primarily of fine-grained minerals. It shows a high level of plasticity over a wide range of water content, and hardens when dried or fired. The large aspect ratio of nanoclay particles corresponds to increased contact area between the polymer and the filler. In addition, it is well known that nano-dimension layered silicate can dramatically change the mechanical and other performances at even very low filler concentration after homogeneous dispersion inside the host matrix [13,43,44]. Figure 2.5 shows the shape and molecular structure of clay used in this research.

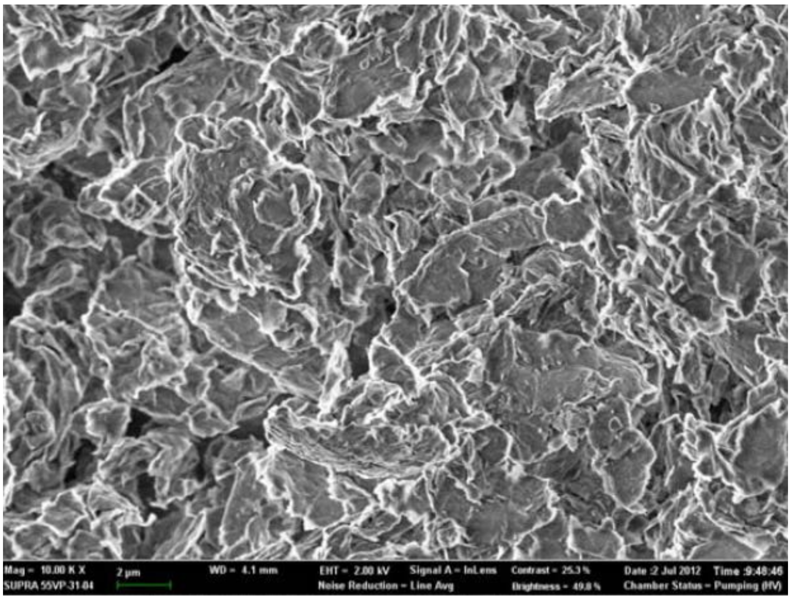


Figure 2.4 FE-SEM image of nanoclay

Figure 2.5 Nanoclay and its molecular structure [44]

2.3.3 Carbon nanotubes

Carbon nanotubes (CNTs) are allotropes of carbon with a cylindrical and one dimensional nanostructure which is shown in Figure 2.6. These cylindrical carbon molecules exhibit extraordinary strength and unique electrical and thermal properties, making them potentially useful in many applications in nanotechnology, electronics, optics, and other fields of materials science. The high electrical conductivity and low specific gravity of CNT can result in excellent mechanical and electrical properties after reinforce to polymer matrix. Furthermore, mixing two or three different materials with CNT together can produce outstanding improvements obviously [45-47]. However, there is no obstacle to the widespread use of CNT as its expensive price has declined annually with mass production increasing recently [15,48,49]. Figure 2.7 shows the CNT used in this research.

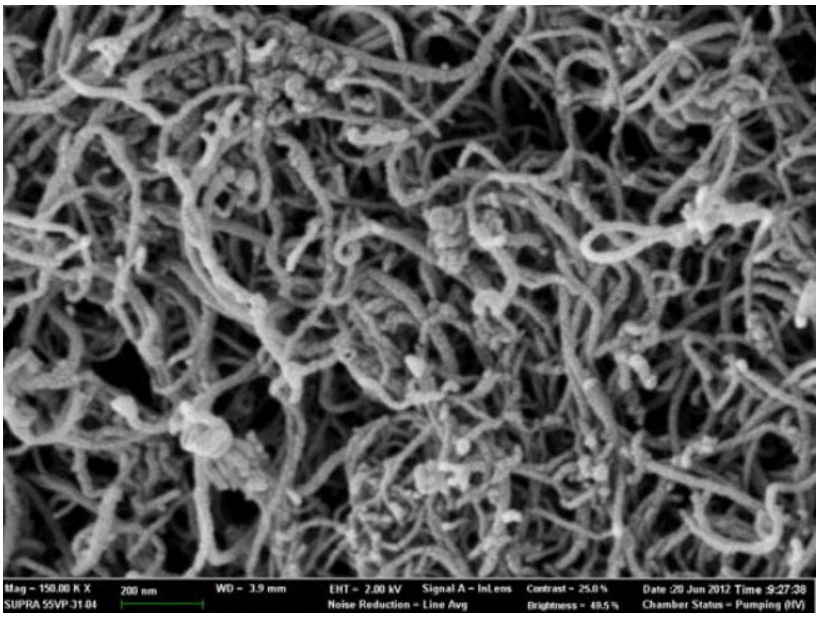


Figure 2.6 FE-SEM image of CNT

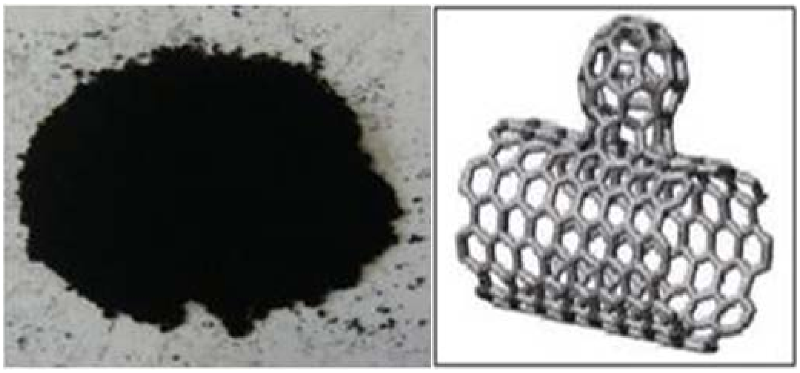


Figure 2.7 CNT and its molecular structure [46]

2.4 Specimens fabrication

2.4.1 Solution blending

The PP was produced by Samsung Company with type HJ400 and used here as the polymer matrix material. Type KR-2000 talc was supplied by the Kyoungki Chemical Company. Nanoclay was produced by Southern Clay Products with type Cloisite 15A and CNTs were obtained from the Hanwha Nanotech Company with type MWNT CM-95. These three kinds of fillers were used as reinforcing materials. Table 2.2 shows the specifications for each filler material[50].

Table 2.2 Filler specifications

Filler	Type	Density (g/cm^3)	Diameter
Talc	KR-2000	2.5~2.8	10μm
Clay	Cloisite 15A	1.66	13nm
CNT	MWNT CM-95	1.8	10-15nm

Melt blending and solution blending are the methods typically used to mix fillers into a polymer matrix. In melt blending, the matrix is held at temperatures above its melting point while the filler is added. In solution blending, the filler is dissolved in the polymer matrix using a certain amount of solvent with temperature below the melting point of this matrix. In this study, xylene (400*ml*) was selected as the optimal solvent because of its low boiling point which can make it relatively easy to remove after mixing process completion. And maleic anhydride (0.1*gram*) was served to accelerate the dissolution of filler into polymer matrix during heating process. Therefore, the matrix polymer could be functionalized and compatible with silicate layers by enhancing its surface energies [24].

Figure 2.8 shows the preparation of PP/clay powders using melt blending and solution blending method. Clay was added at 0.9, 2.9, 4.8, 6.5, 8.2, and 9.9wt%. A hot plate was used to heat this mixture under 135°C with stirring 300 *rpm* for about 2 hours. After heating, this hot mixture was transferred to a beaker containing 400*ml* of ethanol. Then, a Buchner funnel was used to filter this mixture to obtain the precipitate. Finally, this precipitate was dried in an oven at 110°C for about 4 hours to ensure the removal of solvent [50]. The molecular association process of clay platelets into PP matrix by blending method can be shown through Figure 2.9.

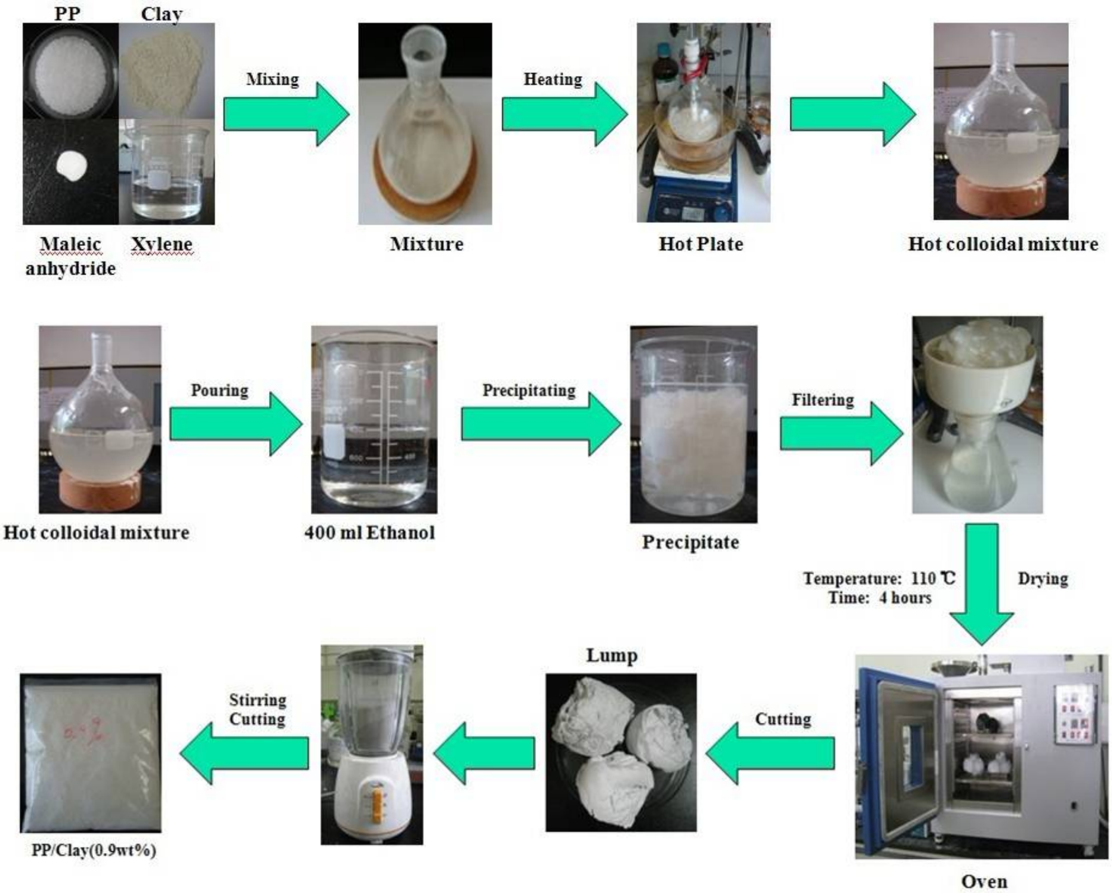


Figure 2.8 Preparation of PP/Clay powders

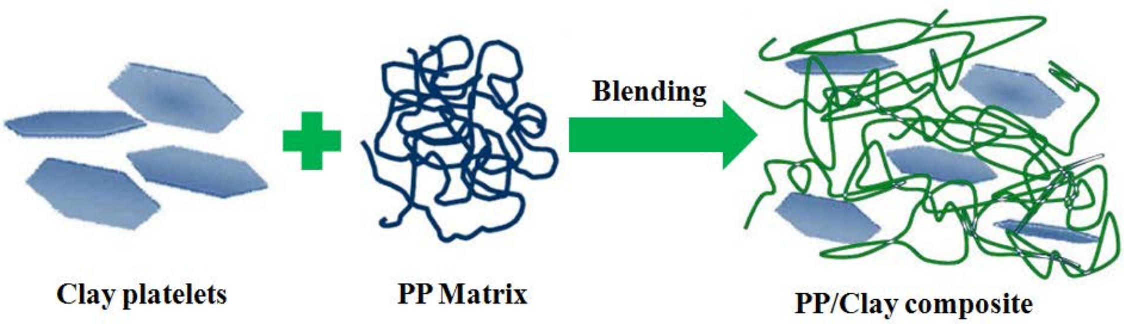


Figure 2.9 Molecular association process by blending method

2.4.2 Specimens fabrication

Composite materials were fabricated in heating press and injection molding machines, as shown in Figure 2.10 and 2.11. The temperature and pressure were fixed at 230°*C* and 35 *MPa* for heating press machine and the curing time for mold was 40 minutes. The square specimens with side length 200*mm* and thickness 3*mm* could be cut into round specimens with diameter 100*mm* through laser cutting machine. An injection molding machine was fixed at 230°*C* for barrel and nozzle temperature and 54.5 *MPa* for injection pressure to make specimens with diameter 29*mm* and thickness 3*mm* [50].

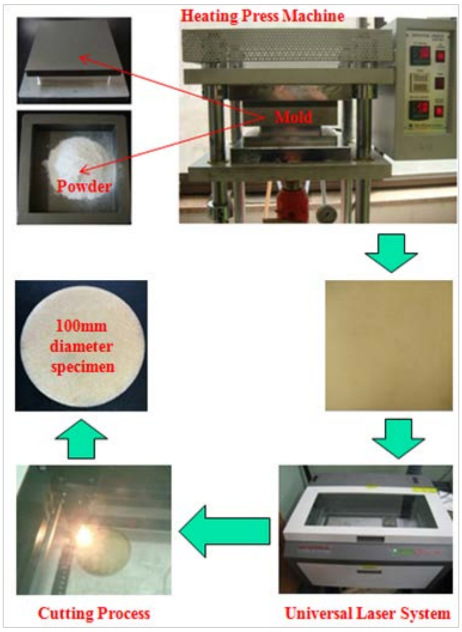


Figure 2.10 Fabrication process of heating press machine

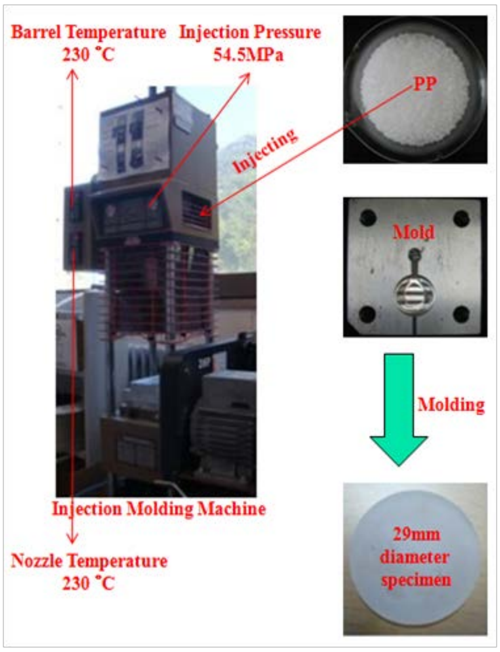


Figure 2.11 Fabrication process of injection molding machine

Chapter 3 Soundproofing properties

3.1 Overview

Soundproofing means reducing the sound pressure with respect to a specified sound source and receptor. There are mainly four kinds of methods for soundproofing property measurement of a give material as shown in Figure 3.1 which are impedance tube test, reverberation chamber test, microphone pulse analyzer test, and sound level meter test. In this research, the specimens for their soundproofing property test is measured by sound TL through impedance tube test and sound pressure level through microphone pulse analyzer test.

Figure 3.1 Soundproofing property test methods

3.2 Impedance tube test

The impedance tubes are specifically designed to measure the main normal indicators of a wide range of acoustical materials. The method is based on ASTM E1050 and ISO 10534-2 standards [51].

In this research, the amplitude of incident and transmitted sound waves was measured with four ¼-inch Brüel & Kjær (B&K) 4196 microphones mounted on a B&K 4206 impedance tube. A B&K 2690 Nexus conditioning amplifier was used to amplify low signals. An HP 35670A frequency analyzer, connected to a computer through a GPIB interface, was used as the sound source and data acquisition device. Program LabView 7.0 would be as the user interface to show the final Sound TL results. The whole setup of impedance tube method was shown in Figure 3.2[50,52,53].

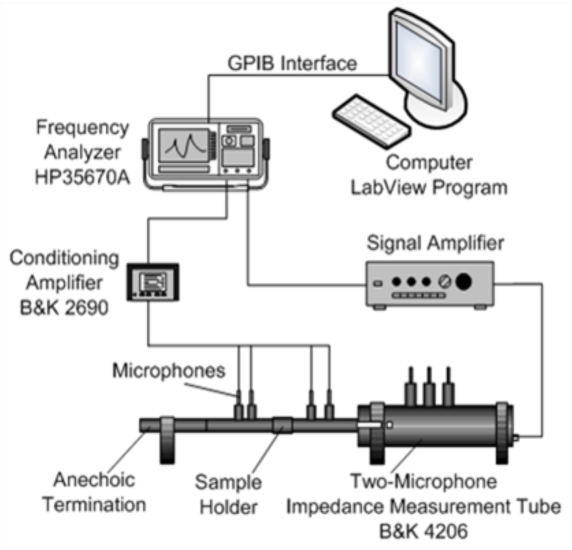


Figure 3.2 Schematic of the four-microphone measurement setup [52]

Figure 3.3 shows each kind of specimen with thickness 3*mm* and diameter 29*mm*. Figure 3.4 shows the specimens for high sound frequency measurements.

Figure 3.3 Impedance tube specimens

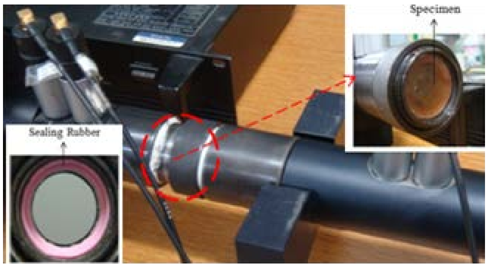


Figure 3.4 High frequency tube test for 29*mm* diameter specimens

3.3 Soundproofing effect results

Table 3.1 shows the specimens used in impedance tube test. All specimens were prepared by talc, clay, CNT and clay/CNT into the PP matrix as function of fillers contents to compare the soundproofing effect.

Table 3.1 Component contents of each kind of specimen

Matrix	Filler	wt%
PP	Talc	95/5 90/10 85/15
PP	Clay	99.1/0.9 95.2/4.8 93.5/6.5
PP	CNT	99.9/0.1 99.5/0.5 99.3/0.7
PP	Clay/CNT	99/0.9/0.1 94.7/4.8/0.5 92.8/6.5/0.7

3.3.1 PP/Talc composites

PP/talc composites were used here as the benchmark against which to compare the performance of PP/Clay and PP/CNT composites [39-41]. Sound TL of PP/talc composites were shown as a function of filler content for 29*mm* specimens. As seen in Figure 3.5, the sound TL values gradually increased with increasing filler content and the PP/Talc(15wt%) composites showed the maximum sound TL with 8 *dB* increasing at 4800 *Hz* compared with pure PP.

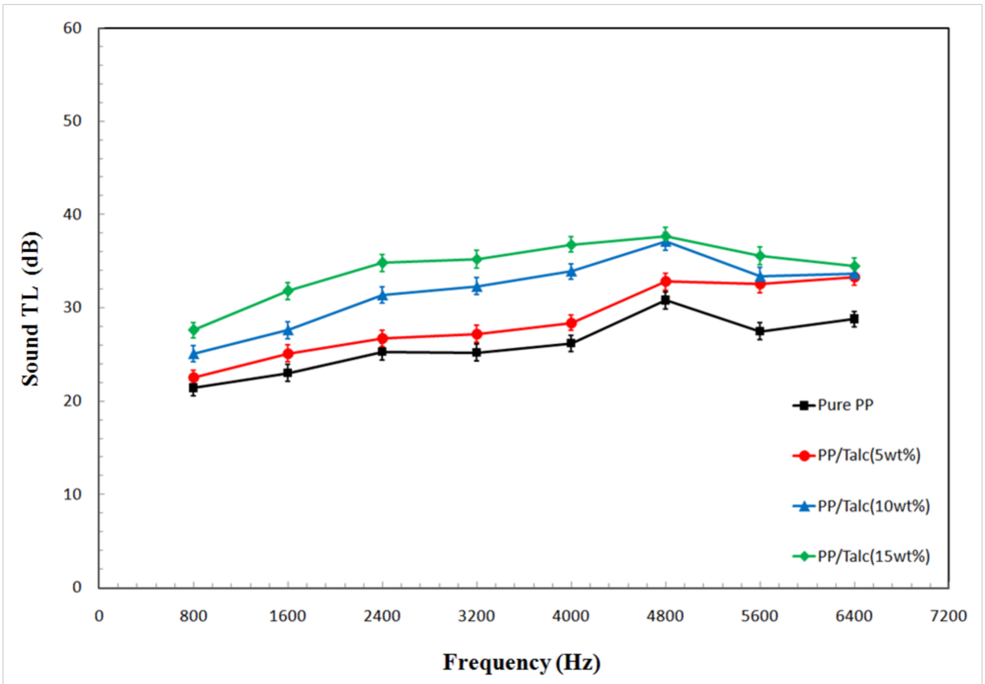


Figure 3.5 Sound TL of PP/Talc composites

3.3.2 PP/Clay composites

Adding nanoclay filler into PP matrix can result in a large contact area available for interaction with the PP matrix because of its laminar structure and high aspect ratio. Therefore, the soundproofing and mechanical properties can be improved at the same time [12-14].

The PP/nanoclay composites showed sound TL values higher than those of the pure PP over the entire sound frequency range along with clay weight percentage increasing. For 29*mm* diameter specimens, PP/Clay(6.5wt%) composite showed the highest sound TL at 4800 *Hz* with 10 *dB* increasing compared with pure PP. Figure 3.6 shows the soundproofing properties of PP/Clay composites as a function of the clay content at high sound frequencies.

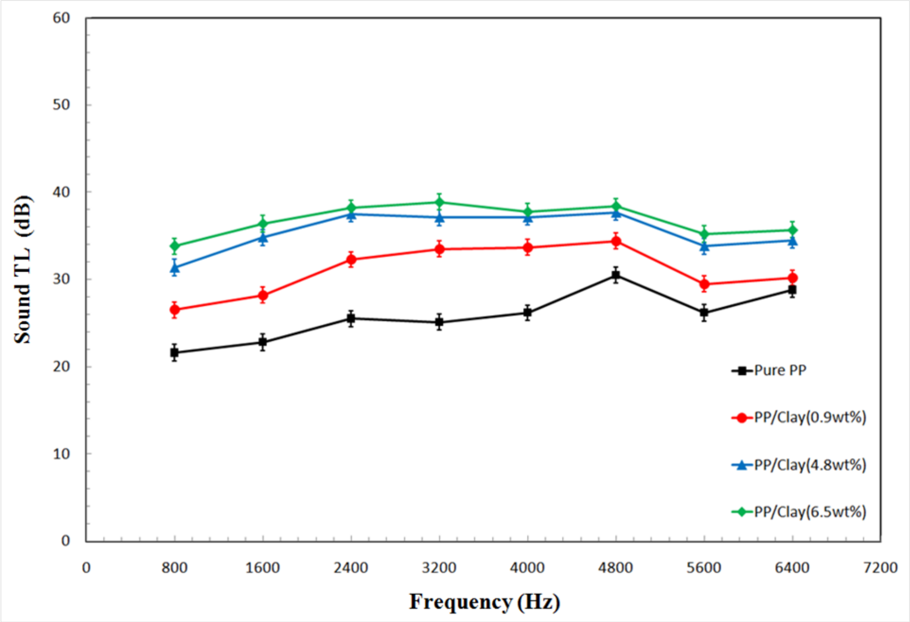


Figure 3.6 Sound TL of PP/Clay composites

As PP/Clay(6.5wt%) composite showed the best soundproofing effect among all PP/Clay composites, the relationship between soundproofing ability and the thickness of the specimen was shown in Figure 3.7 with pure PP and 29*mm* diameter specimens of PP/Clay(6.5wt%) composite. The sound TL increased with sound frequency and thickness of the specimens [50,54].

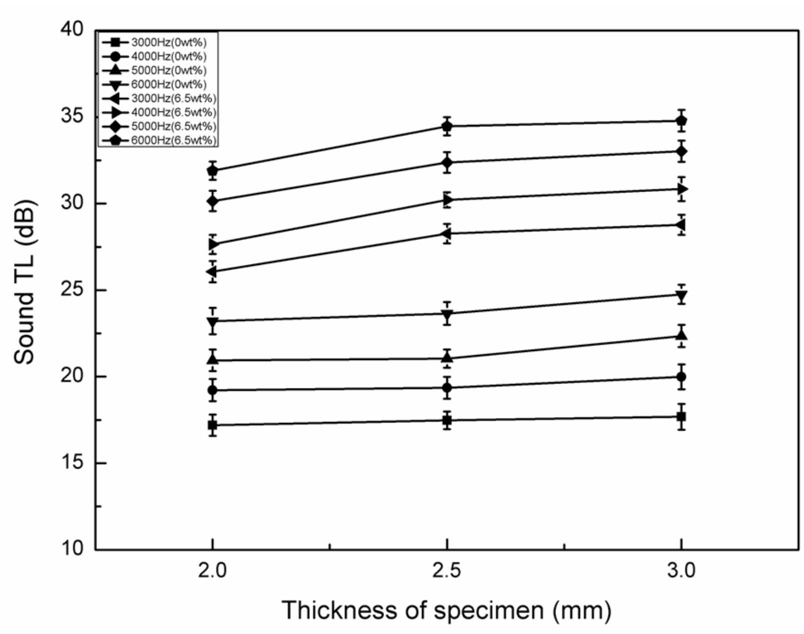


Figure 3.7 Comparison of sound TL as a function of thickness and clay content

3.3.3 PP/CNT composites

Even very low CNT loading reinforced polymer matrix can yield remarkable mechanical, electrical, and thermal properties of PP/CNT composites according to lots of recent research [15,46-48,55].

Figure 3.8 shows the soundproofing properties of PP/CNT composites as a function of the CNT content at high sound frequencies.

All the specimens containing CNT showed better soundproofing properties than those of the pure PP. For 29*mm* specimens tested in high sound frequency range, the sound TL values gradually increased with increasing filler content until 4800*Hz* and the highest sound TL was found on PP/CNT(0.7wt%) composite with 5 *dB* increasing at 4800 *Hz* compared with pure PP. It demonstrated that very low loading of CNT into the PP matrix could improve the soundproofing properties much.

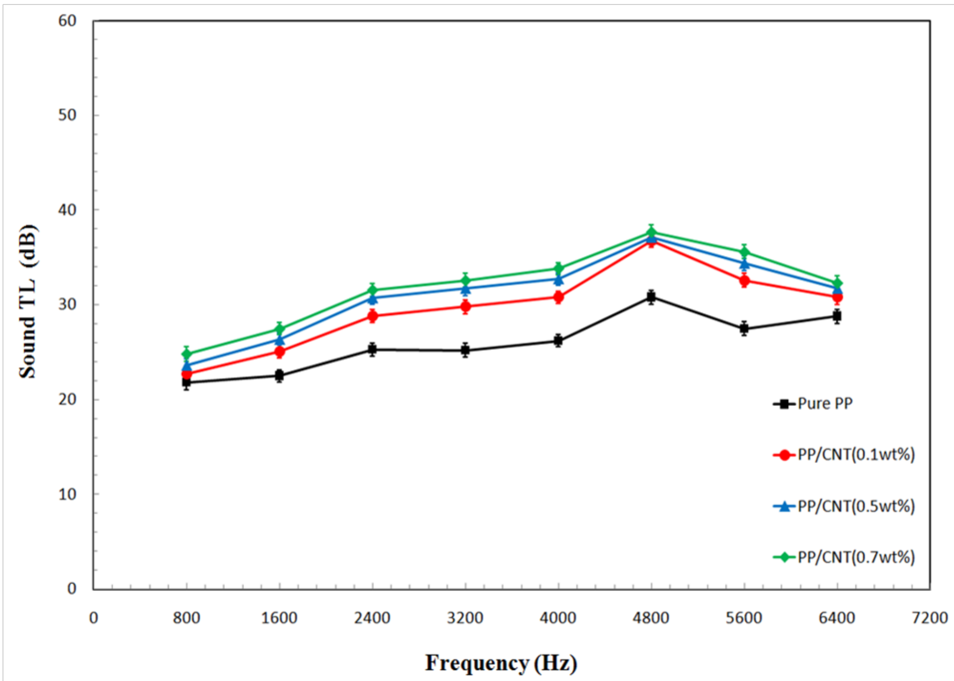


Figure 3.8 Sound TL of PP/CNT composites

3.3.4 PP/Clay/CNT composites

The soundproofing effect of the nanoclay/CNT combination was higher than the effect of either component alone with 0.1~0.7wt% CNT and 0.9~6.5wt% nanoclay containing. The soundproofing effects of these PP/Clay/CNT composites are shown as a function of filler content with high sound frequencies in Figure 3.9.

All the specimens containing both clay and CNT showed better soundproofing properties than those of the pure PP. Sound TL increased with more filler loading gradually. The highest sound TL was found on PP/Clay(4.8wt%)/CNT(0.5wt%) composite with 20 *dB* increasing at 4800 *Hz* compared with pure PP. Moreover, this kind of PP/Clay(4.8wt%)/CNT(0.5wt%) composite also showed perfect soundproofing effect than PP/Clay(6.5wt%) composite and PP/CNT(0.7wt%) composite over all sound frequency range.

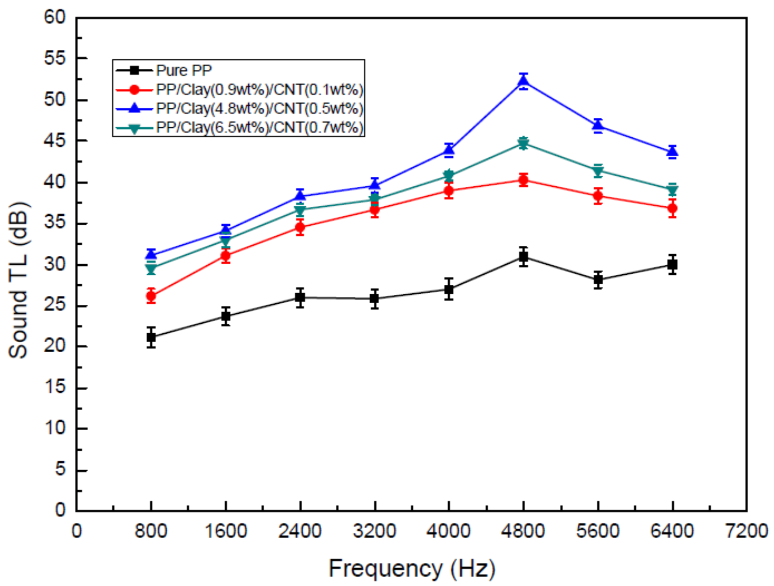


Figure 3.9 Sound TL of PP/Clay/CNT composites

3.3.5 Sound TL comparison

The soundproofing properties of these PP/Talc, PP/Clay, PP/CNT, and PP/Clay/CNT composites were compared with those of pure PP as a function of filler content. As PP/Talc(15wt%), PP/CNT(0.7wt%), PP/Clay(6.5wt%), and PP/Clay(4.8wt%)/CNT(0.5wt%) composites had the largest sound TL among each kind of filler loading group, a comparison including all composites above were shown with high sound frequencies in Figure 3.10 which represented the highest soundproofing effect.

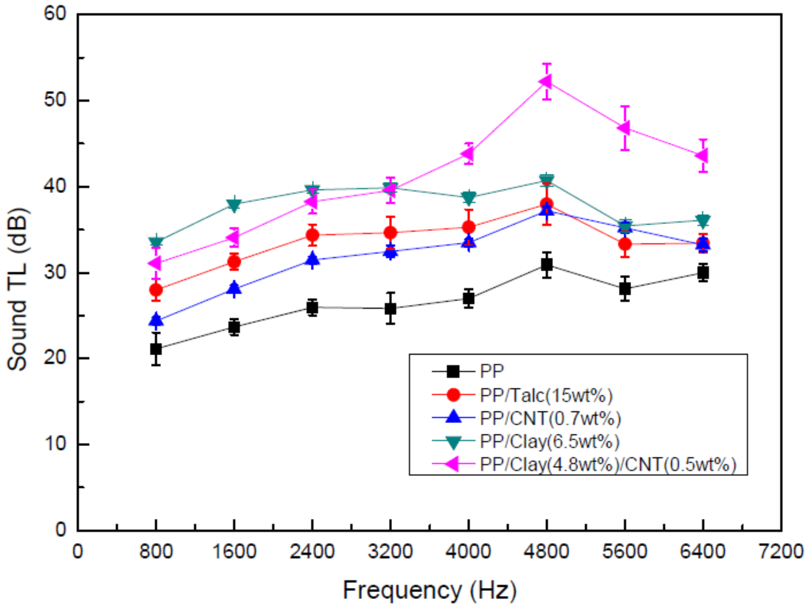


Figure 3.10 Comparison of the sound TL values of pure PP, PP/Talc, PP/Clay, PP/CNT, and PP/Clay/CNT composites

All specimens exhibited better soundproofing properties than pure PP matrix. In particular, PP/Clay(4.8wt%)/CNT(0.5wt%) composite showed remarkable maximum Sound TL of 15~20 *dB* at 3200~6400 *Hz* which exhibits that the PP/Clay(4.8wt%)/CNT(0.5wt%) composite has the best soundproofing property.

3.4 Microphone pulse analyzer test

The sound pressure level of a given material can be measured through microphone pulse analyzer test. In this research, 100*mm* diameter specimens of pure PP, PP/Clay(6.5wt%), PP/CNT(0.7wt%), PP/Clay(4.8wt%)/CNT(0.5wt%) composites were tested for their soundproofing property by sound pressure level measurement. Microphone 4189 with sensitivity 50 *mV/Pa*, frequency 6.3 *Hz*~20 *KHz*, dynamic range 14.6~146 *dB*, temperature -30~150℃ and pulse analyzer 3560C with 2 modules, 17 inputs, 3 generator output channels were used here for soundproofing property test as shown in Figure 3.11. Moreover, the test condition was set with temperature 16℃, humidity 20%, distance 100*mm* and vertical direction between microphone and specimens.

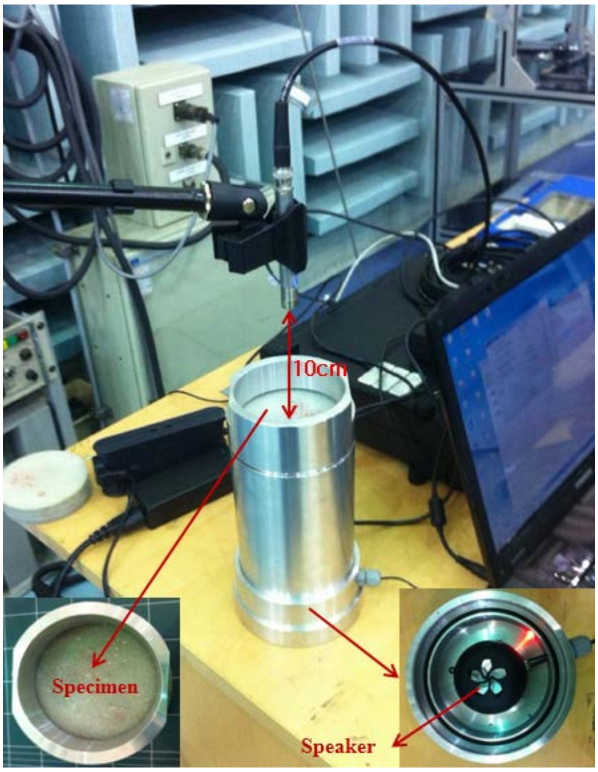


Figure 3.11 Microphone pulse analyzer test

In order to prevent sound leakage, a special specimen holding tube was designed as shown in Figure 3.12. Therefore, the final measurement results can be more precise and believable.

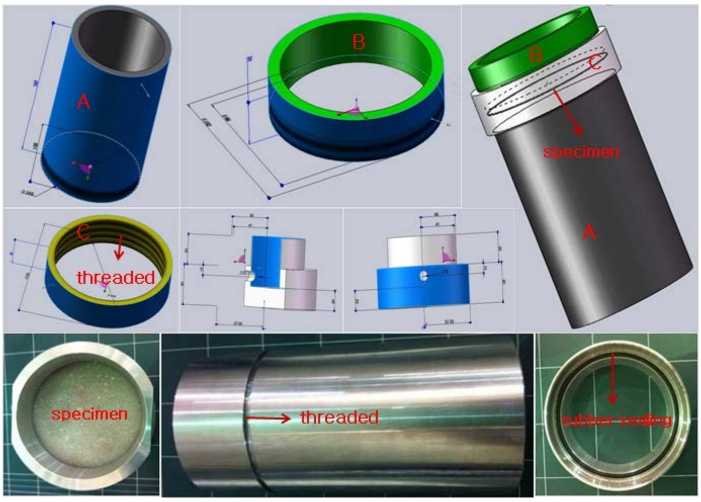


Figure 3.12 Design of specimen holding tube

Figure 3.13 shows the results of microphone pulse analyzer for 100*mm* diameter specimens of pure PP, PP/Talc(15wt%), PP/Clay(6.5wt%), PP/CNT(0.7wt%), and PP/Clay(4.8wt%)/CNT(0.5wt%) composites at 1000 *Hz*, 3000 *Hz*, 5000 *Hz*, and 7000 *Hz*, respectively.

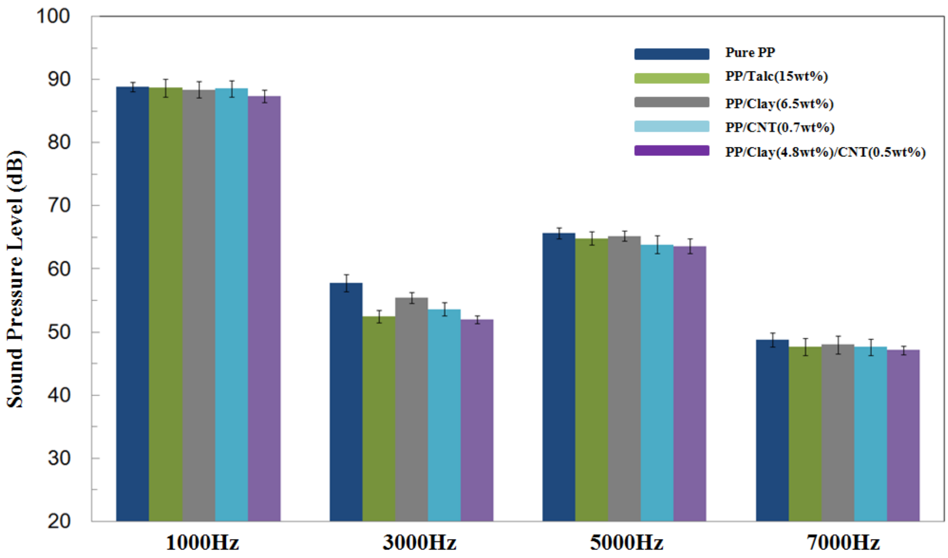


Figure 3.13 Microphone pulse analyzer test at 1000, 3000, 5000, and 7000 *Hz*

3.5 Blind test

A blind test is a scientific experiment where some of the people involved are prevented from knowing certain information that might lead to conscious or subconscious bias on their part, invalidating the results. The objective of this research is to investigate which kind of nanocomposite shows the best soundproofing property by response of the ear in order to verify and prove the correctness of experiment results.

The test was set with temperature 23℃, humidity 16% in a classroom and included 10 participants into two groups. Hearing the sound coming out under the specimens which were assembled tightly in the specimen holding tube, the participants would make a decision to choose a larger sound when they heard after sound ringing. As PP/Clay(4.8wt%)/CNT(0.5wt%) composites showed the best soundproofing property among all the nanocomposites specimens, a test between pure PP and PP/Clay(4.8wt%)/CNT(0.5wt%) composites was compared in this blind test with sound frequency at 3000 *Hz*, 5000 *Hz*, and 7000 *Hz*, respectively. As seen in Figure 3.14, many people choose pure PP as they can hear much larger sound. Therefore, it verifies that PP/Clay(4.8wt%)/CNT(0.5wt%) composites shows better soundproofing property than pure PP.

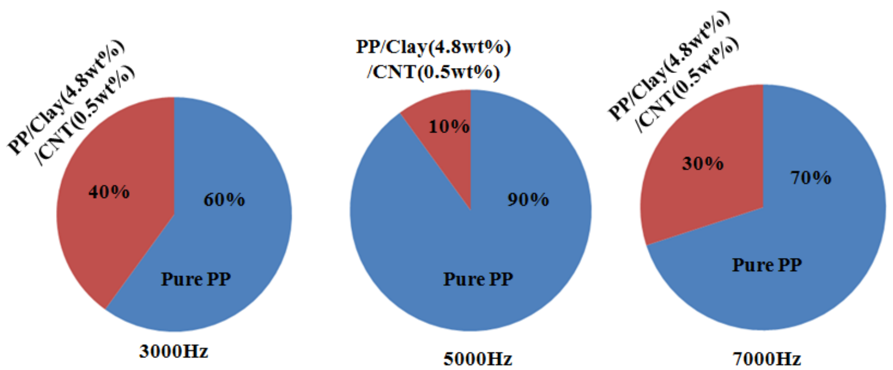


Figure 3.14 Results of blind test

Chapter 4 Mechanical properties

4.1 Overview

Elementary and important mechanical properties include density, Young's modulus, elongation, and Poisson's ratio which show inherent characteristic such as elasticity, plasticity, strength, hardness, and ductility of the composite materials. It is vital and necessary to test mechanical properties of one kind of new composite materials before real items production and application [56-60]. Many researchers have studied mechanical properties of nanocomposites with various fillers reinforced polymers[61-65]. Kristiina Oksman *et al*. studied the mechanical properties and morphology of impact modified polypropylene-wood flour composites [66]. Yuanqing Xiang *et al*. studied the tensile mechanical properties of a new polymer/clay nanocomposite [67]. Adding clay into pure PP can change the inner crystal structure of PP and alter its mechanical properties. The following sections discuss the parameters that yield these changes.

4.2 Measurement devices

The ASTM standard method (D638-03) was used here for mechanical properties evaluation. Figure 4.1 and Table 4.1 show the shape and dimensions of the samples made for tensile test. A universal material testing machine (Lloyd Instruments LR50K) and strain indicator (NI SCXI-1520, 8-channel universal strain/bridge) were used for the tensile and strain tests, each operating at 5 mm/min with a preload of 5.0 *N*[50]. The test bed is shown in Figure 4.2.

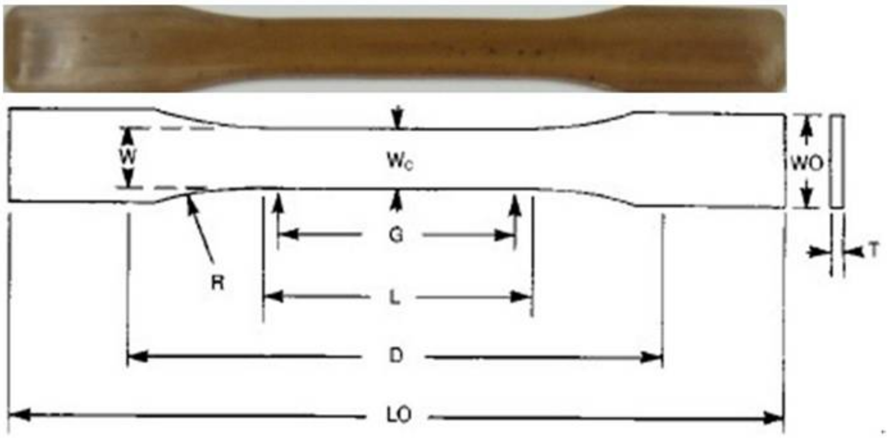


Figure 4.1 Specimen for tensile test [50]

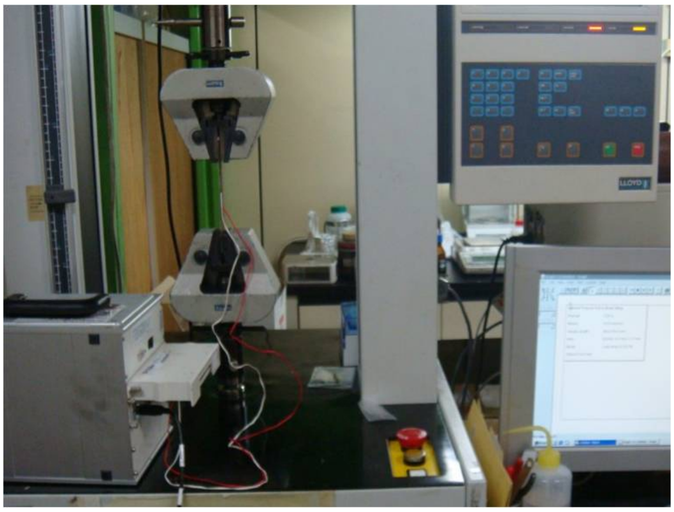


Figure 4.2 Universal material testing machine for tensile test

Table 4.1 Dimension and value of specimen for tensile test [50]

Dimensions	Values(mm)
W(width of narrow section)	13
L (length of narrow section)	60
WO(width overall)	19.55
LO (length overall)	178
G (gauge length)	50
D (distance between grips)	100
R (radius of fillet)	76
T (thickness)	3.45

4.3 Measurement results

Young's modulus, also known as the tensile modulus or elastic modulus, is a measure of the stiffness of an elastic material and is a quantity used to characterize materials. Young's modulus is the most common elastic modulus, sometimes called the modulus of elasticity. Elongation shows plastic deformation ability of the materials and Poisson’s ration is an elastic constant which shows transverse deformation ability of the materials [50].

4.3.1 Density

Figure 4.3 showed a density comparison of pure PP and nanoclay-reinforced PP composites. In general, the trend of density was inclined toward linear increase from 0 to 9.9wt% specimens. The 4.8wt% and 6.5wt% clay reinforced PP composites corresponded to 16.7% and 24.9% density increase compared to pure PP, respectively. As the density of clay is larger than that of pure PP, a small amount of clay reinforced PP can enhance the density of these PP/Clay composites. Further, the density in semi-crystalline PP based nanocomposites can greatly affect the matrix morphology by the addition of coupling agent and usually leads to good clay intercalation instead of exfoliation.

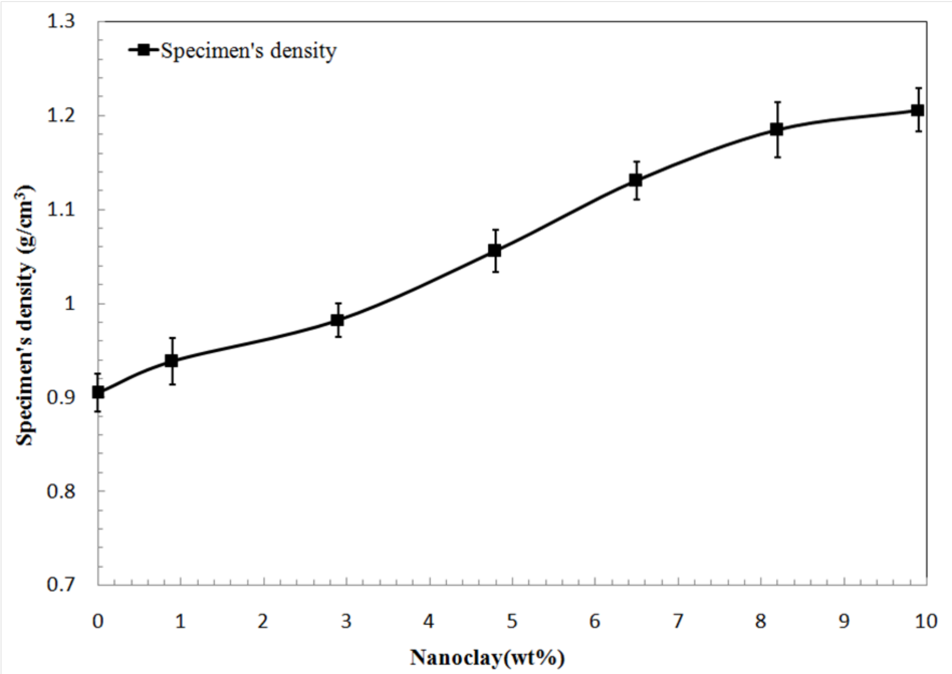


Figure 4.3 Density comparison between PP/Clay composites and pure PP

4.3.2 Elongation

Figure 4.4 shows that the elongation of these composites will increase from 0 to 9.9wt%. It means that more clay filler specimens will be broken slowly during tensile test and have better plastic property. Layered silicate reinforcement of polymer composites is known for improving mechanical performance of resulting hybrids up to a certain level, more particularly when distributed homogenously inside host matrix and present in a state where order of the clay platelets are completely destroyed and polymer chains are randomly quenched between the silicate layers.

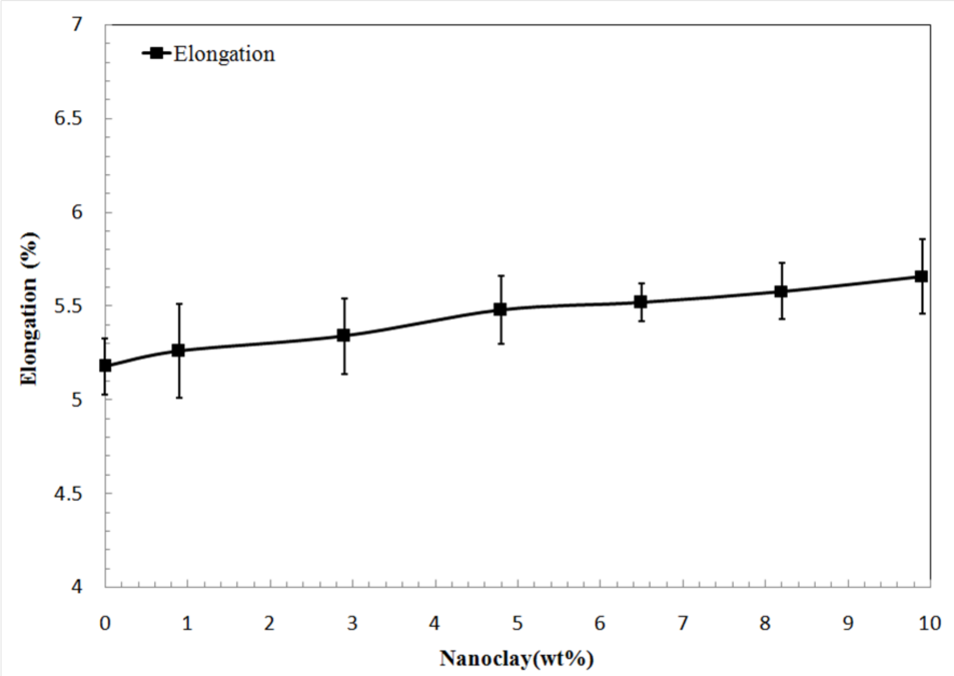


Figure 4.4 Elongation of PP/Clay composites

4.3.3 Poisson's ratio

Figure 4.5 shows that the Poisson's ratio is a little increased following the clay filler increasing. It means that the lateral deformation of this nanoclay reinforced PP composite is not so remarkable. The layer separation, especially exfoliation, depends on the establishment of favorable interactions between the polymer and the clay surface and the subsequent system energy reduction.

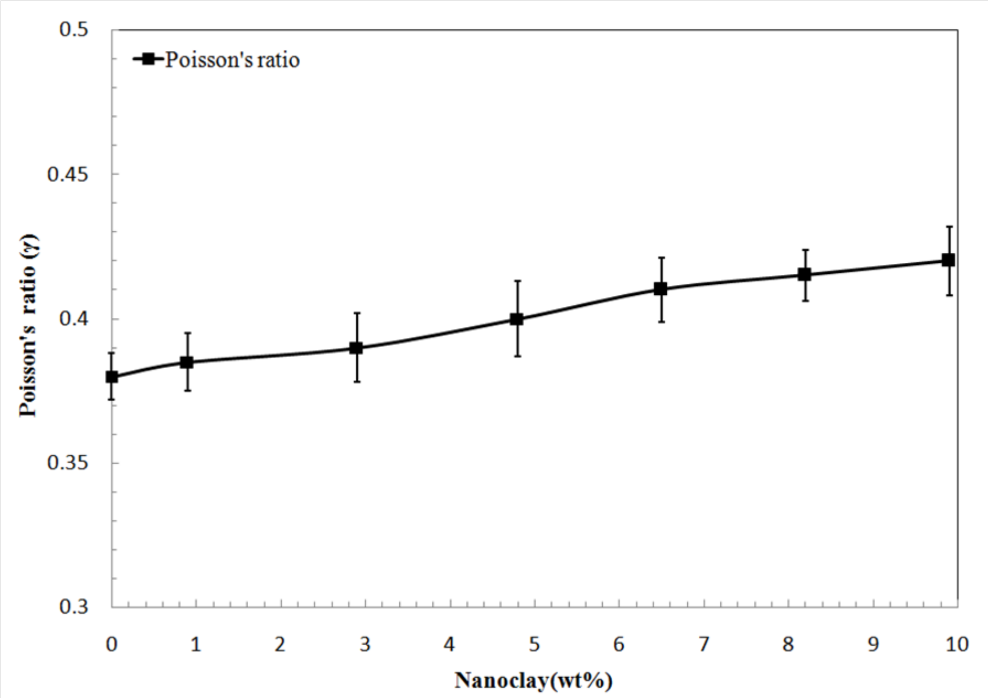


Figure 4.5 Poisson’s ratio of PP/Clay composites

4.3.4 Stress-strain curve

Stress-strain curve is unique for each material and is found by recording the amount of deformation at distinct intervals of tensile or compressive loading. These curves reveal many of the properties of a material such as ultimate strength and yield strength. The stress-strain curves for all specimens: pure PP, PP/Clay(0.9wt%), PP/Clay(2.9wt%), PP/Clay(4.8wt%), PP/Clay(6.5wt%), PP/Clay(8.2wt%), and PP/Clay(9.9wt%) are shown in Figure 4.6. And the ultimate strength of PP/Clay(0.9wt%), PP/Clay(2.9wt%), PP/Clay(4.8wt%), PP/Clay(6.5wt%), PP/Clay(8.2wt%), and PP/Clay(9.9wt%) increases 2.5%, 11.8%, 19.3%, 28.5%, 34.8%, and 41.1%, respectively.

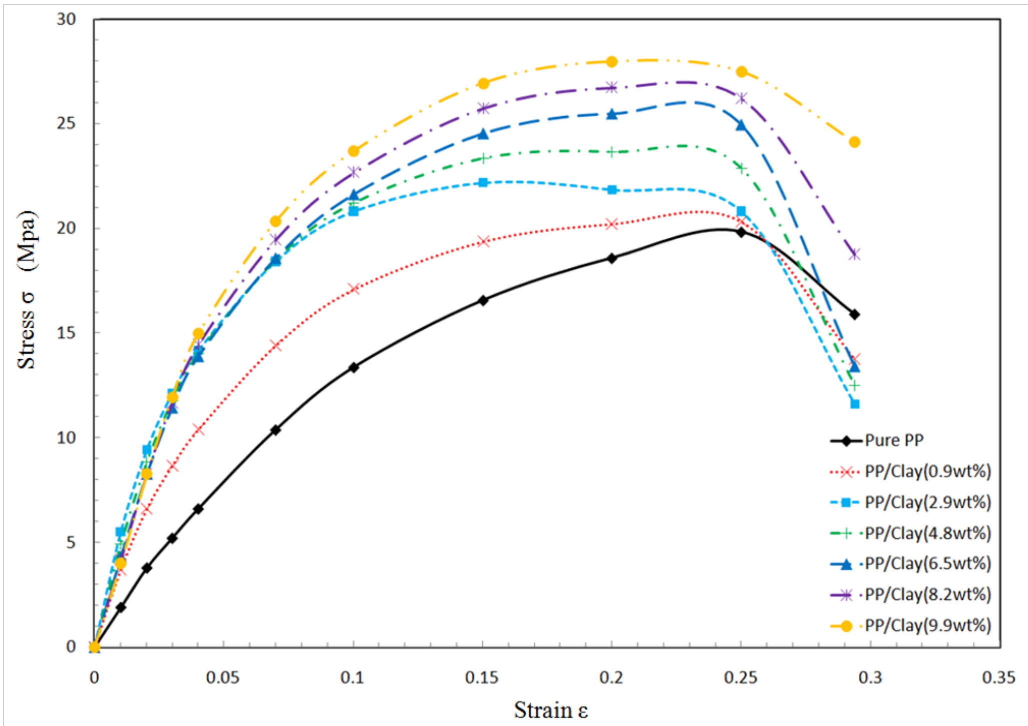


Figure 4.6 Stress-strain curve for PP/Clay composites

4.3.5 Dynamic mechanical properties

The mechanical behaviors of polymers and polymer composites are commonly studied by dynamic mechanical analysis (DMA) which includes measurement of storage modulus (M′) and loss modulus (M″) of the specimens under oscillating load against time, temperature, or frequency of oscillation. Storage modulus shows the elastic property and loss modulus shows the plastic property of composite materials. The ratio M″/M′ is the loss tangent (tanδ) which shows the viscoelasticity ratio of the given composite materials.

The DMA experiment has been done in 3 point bending method by using dynamic mechanical thermal analyzer machine which was made in Mettler Toledo Company, Switzerland, 2010 with type of DMA/SDTA861e, temperature range -150~500 ℃, frequency range 0.001~1000*Hz*, force range 0.005~40*N*, heating scan rate 0.1~20 ℃*/min*, and cooling scan rate 0.1~10 ℃*/min*. The size of specimen for DMA test is 60*mm*, 8*mm*, and 3*mm* for length, width, and thickness, respectively, as shown in Figure 4.7. Figure 4.8 shows the process of DMA experiment and Figure 4.9 to 4.11 show storage modulus, loss modulus, and tanδ changing with temperature, respectively. The best dynamic mechanical property is found on PP/Clay(9.9wt%) composite with highest storage modulus which shows the best elastic property.

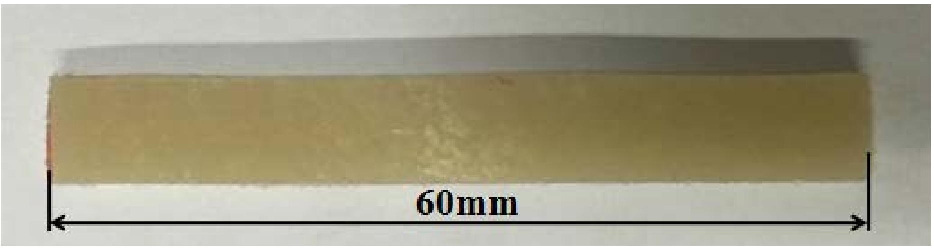


Figure 4.7 PP/Clay(0.9wt%) specimen for DMA experiment

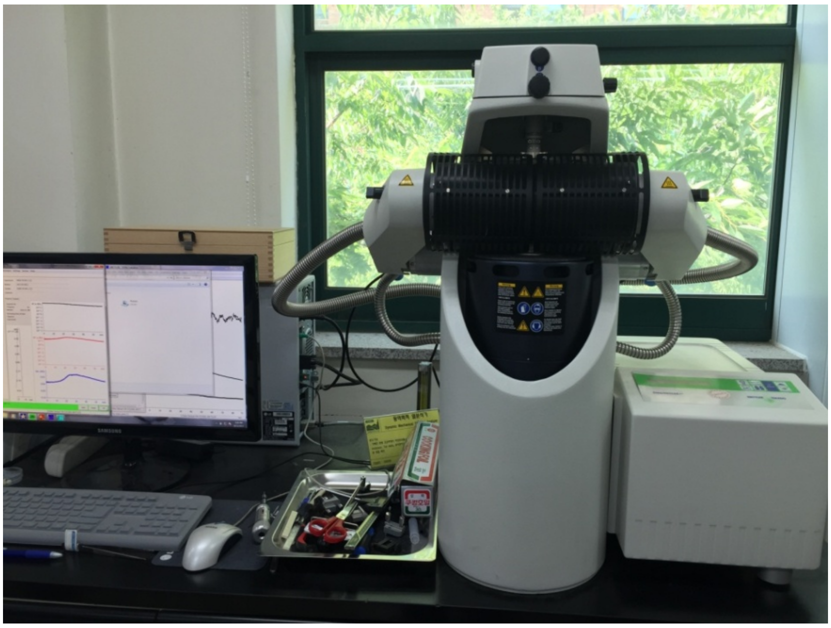


Figure 4.8 Setup for DMA experiment

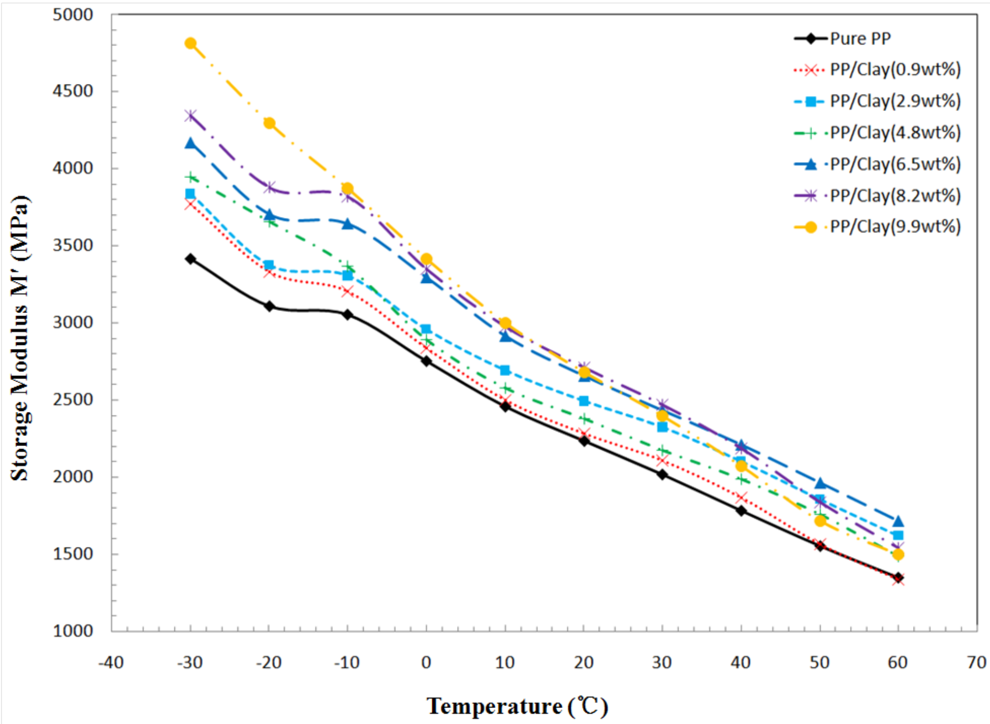


Figure 4.9　Storage modulus (M′) changing with temperature

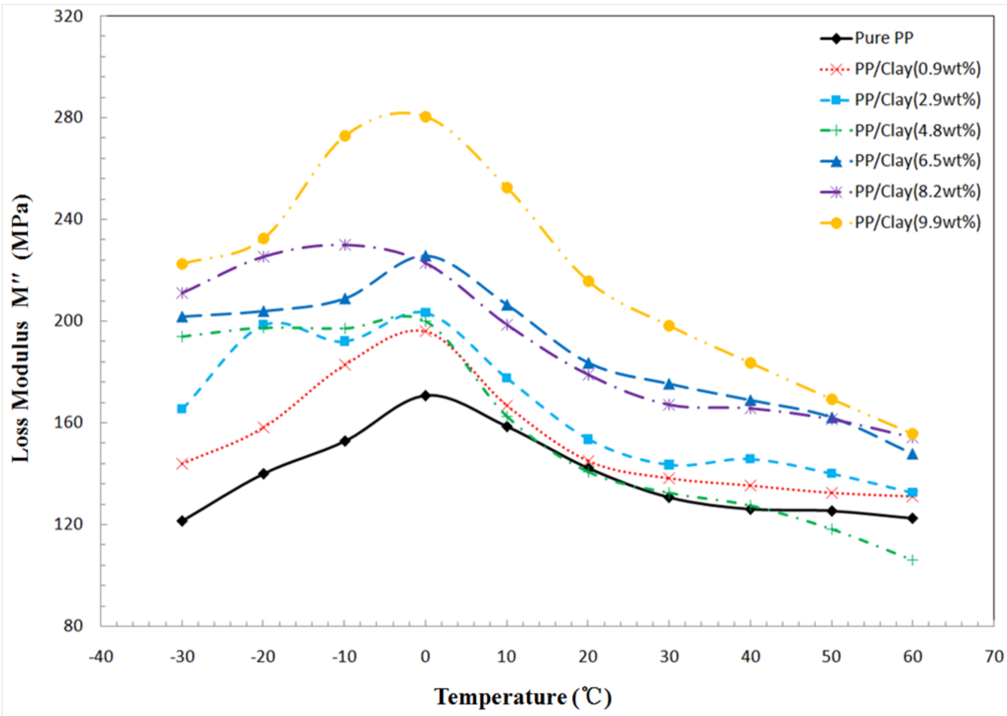


Figure 4.10　Loss modulus (M″) changing with temperature

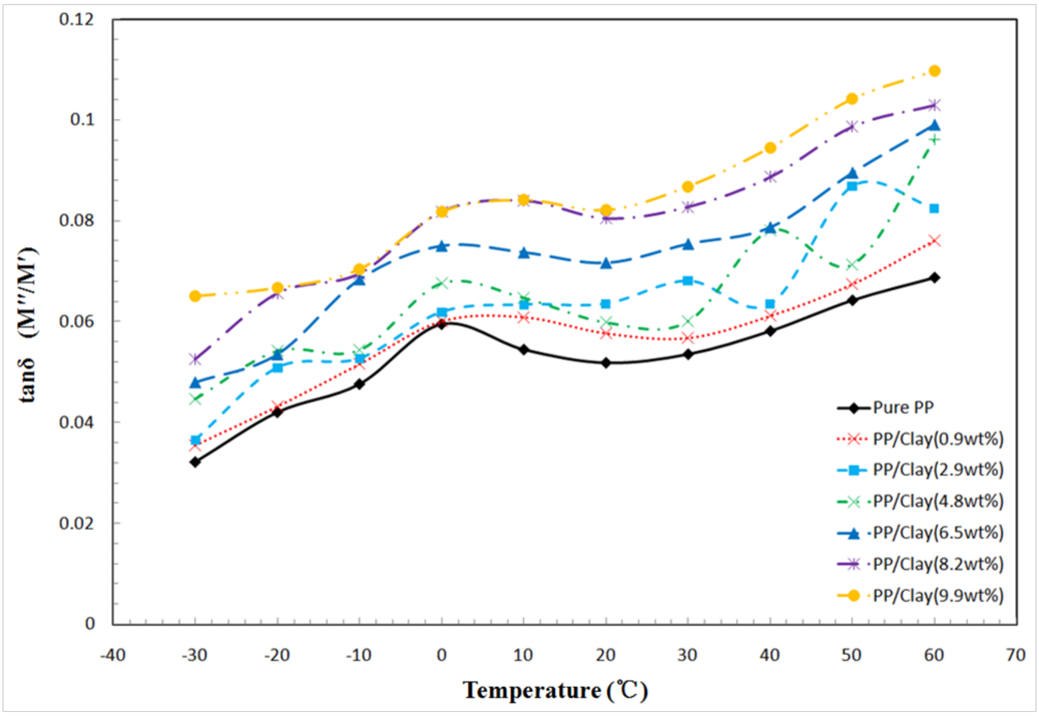


Figure 4.11 Tanδ (M″/M′) changing with temperature

4.4 Growth of mechanical properties with clay increasing

As shown in Figure 4.3 to Figure 4.11, along with clay weight percentage increasing from 0 to 6.5wt%, the mechanical properties including density, elongation, Poisson's ratio, ultimate strength, storage modulus, and loss modulus are increasing linearly. The actual values are shown in Table 4.2.

Table 4.2 Linear growth for mechanical properties with clay increasing

Mechanical property	Rate of increase compared with pure PP		
	PP/Clay(0.9wt%)	PP/Clay(4.8wt%)	PP/Clay(6.5wt%)
Density	1.2%	2.4%	2.9%
Elongation	3.8%	9.6%	15.4%
Poisson's ratio	1.3%	7.9%	10.5%
Ultimate strength	20.5%	39.1%	52.9%
Storage mudulus	10.5%	27.3%	41%
Loss mudulus	14.7%	30.4%	64.1%

Chapter 5 Synergistic effects in nanocomposites

5.1 Overview

Improvements on soundproofing and mechanical properties of nanocomposites are due to a synergistic effect between the matrix and the filler. As the filler disperses homogeneously within the matrix, the three-dimensional structure formed between the filler particles themselves or between the filler particles and the matrix can enhance the synergistic effect which can give a rise to various properties not present in base materials. These synergistic effects have been studied by numerous researchers in a variety of fields such as the mechanical characteristics of multi-graphene platelets (MGPs) and the electrical conductivity of graphite nanoplatelets/CNT epoxy composites[17,45,69-71].

Nowadays, a variety of materials and composites are synthesized to improve the properties of traditional materials depending on synergistic effects which arise from the structural characteristics, the distribution status of the filler in the matrix, and fabrication conditions [72-78]. Figure 5.1 shows the synergistic effect with filler of clay and CNT reinforced the matrix material together. The internal microstructure of these nanocomposites can be observed by transmission electron microscope (TEM). Figure 5.2 shows the TEM image of pure PP.

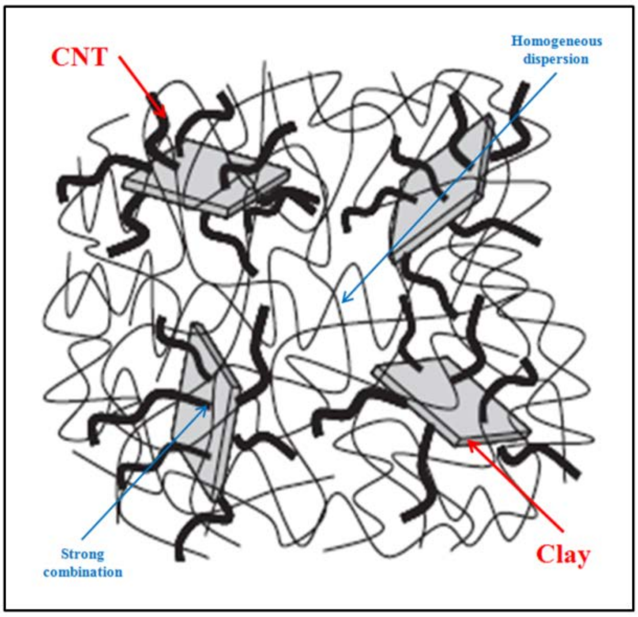


Figure 5.1 Synergistic effect between the filler and the matrix

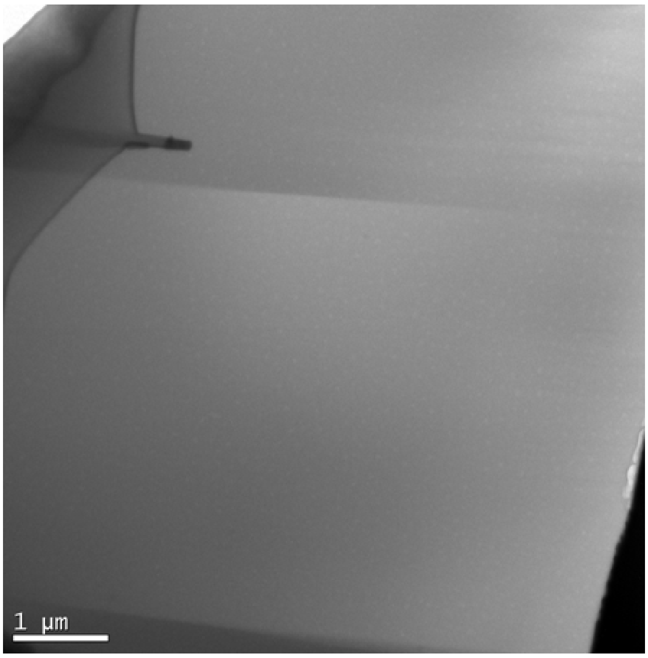


Figure 5.2 TEM image of pure PP

5.2 Synergistic effects on soundproofing property

The uniform dispersion of filler such as clay or CNT in PP matrix can improve mechanical, electrical and acoustic properties of the pure PP.

Figure 5.3 shows TEM micrographs of a PP/Clay (6.5wt%) composite. The two-dimensional, black, plate-like clay particles are homogenously dispersed in PP matrix. And PP/Clay(6.5wt%) composite showed the best soundproofing effect with 15*dB* and 40*dB* higher than that of pure PP at low and high frequencies range, respectively.

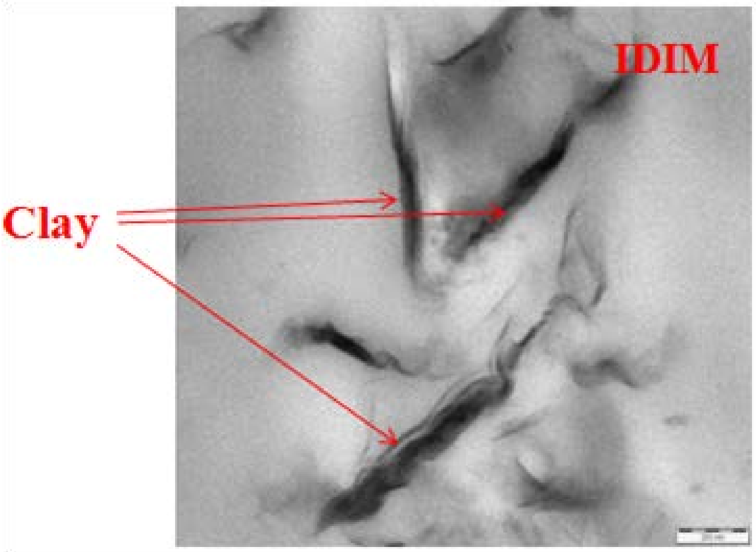


Figure 5.3 TEM image of PP/Clay(6.5wt%) composite

The incident sound will reflect as colliding with clay particles when transfer through the PP matrix. Due to different dispersion and size of clay particles, the incident sound will reflect many times with different amount each time. The reflection energy is contributed by the reflections on the material surface and interfaces between layers and between clay particles and the matrix. The clay particles have a multi-reflection effect contributing to the reflection energy. Such reflection plays a major role in enhancing soundproofing ability. The absorption energy is mainly determined by clay particles in PP matrix due to its viscoelasticity and the inner fraction on interfaces between clay and matrix interfaces. However, too much weight percentage content of clay will form hard segments and result in the decrease of the viscoelasticity which can reduce absorption ability for sound waves. For clay reinforced PP matrix, 6.5wt% will be the optimal amount for clay to reinforce PP compared with weigh percentage of clay filler. Figure 5.4 shows the detour propagation of sound wave in PP/Clay composite.

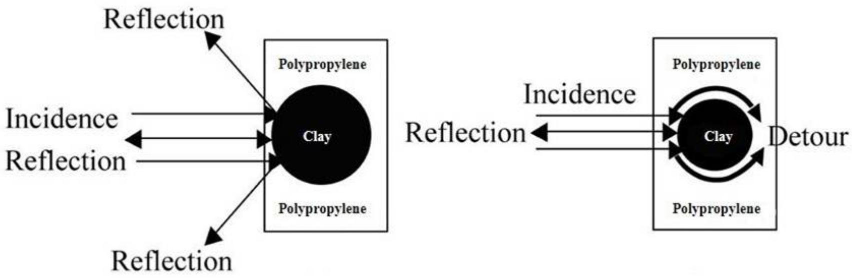


Figure 5.4 Detour propagation of sound wave in PP/Clay composite

As discussed above, these PP/Clay composite specimens exhibited superior soundproofing properties due to the conversion of sound energy to heat which was dissipated by damping effects caused by sound wave transmission and vibrations in the small gaps between the clay particles and the PP matrix. The addition of inorganic fillers to polymeric materials can change the soundproofing characteristics of the material depending on the mass of the filler and its dispersion in the polymer matrix. Normally, this phenomenon may be due to changes in the microstructure of the polymer. Sound TL can be determined by the inner crystal grain size, which is the average distance between crystal grains and deformations caused by sound wave transmission [79]. However, more clay particles over a threshold value may move to together and form aggregation which can prevent sound transmission as shown in Figure 5.5.

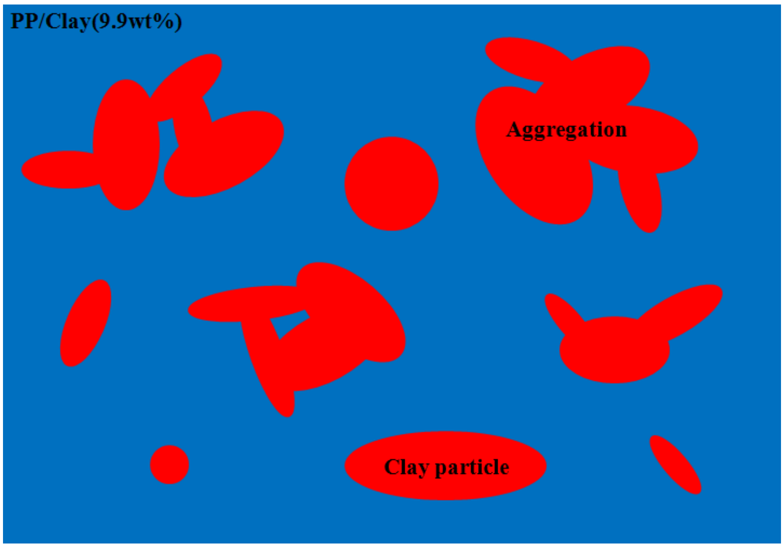


Figure 5.5 Aggregation formed by clay particles in PP matrix

Table 5.1 shows sound TL values of pure PP and PP composites containing various fillers. All specimens were measured at identical frequencies both in low and high frequency regions. The PP/Clay/CNT composites exhibited greater soundproofing effect than that of pure PP, PP/Clay, and PP/CNT composites. This enhancement is mainly due to synergistic effects between these two types of filler material reinforced PP, simultaneity.

Table 5.1 Comparison of sound TL for each nanocomposite

Specimen	Frequency	Sound TL	Frequency	Sound TL
PP	520 *Hz*	12.0 *dB*	4800 *Hz*	29.5 *dB*
PP/Talc(15wt%)	520 *Hz*	15.4 *dB*	4800 *Hz*	37.0 *dB*
PP/Clay(6.5wt%)	520 *Hz*	15.4 *dB*	4800 *Hz*	41.2 *dB*
PP/CNT(0.7wt%)	520 *Hz*	20.0 *dB*	4800 *Hz*	36.5 *dB*
PP/Clay(4.8wt%)/CNT(0.5wt%)	520 *Hz*	25.0 *dB*	4800 *Hz*	51.5 *dB*

Figure 5.6 shows a TEM micrograph of a PP/Clay(4.8wt%)/CNT(0.5wt%) composite. The PP chains are entangled and wrapped around the one-dimensional structure of CNT and two-dimensional structure of clay platelets for PP/Clay/CNT composite. The interaction between PP chains, CNT and clay platelets is much stronger than that of PP chains and Clay or CNT alone, which can prevent and block more sound wave transmission and reflection through such complicated medium. Therefore, more sound energy will be changed into heat energy during this process and more sound TL will be achieved. Figure 5.7 shows sound wave transmission through PP/Clay/CNT composite. As incident sound propagates into the specimens from outside, the incident energy is transformed into either reflected or transmitted sound by interactions and reflection with homogeneously dispersed clay, CNT, and clay/CNT hybrid particles. However, composites with three-dimensional structures between two or more fillers showed higher soundproofing effects than those composites containing only one tpye of filler, such as PP/Clay composite or PP/CNT composite. Seen from Table 5.1, the sound TL for PP/Clay/CNT composite is about 5~10 *dB* more than PP/CNT composite and PP/Clay composite at low sound frequency ranges and about 10~15 *dB* more than PP/Clay composite and PP/CNT composite at high sound frequency ranges[24].

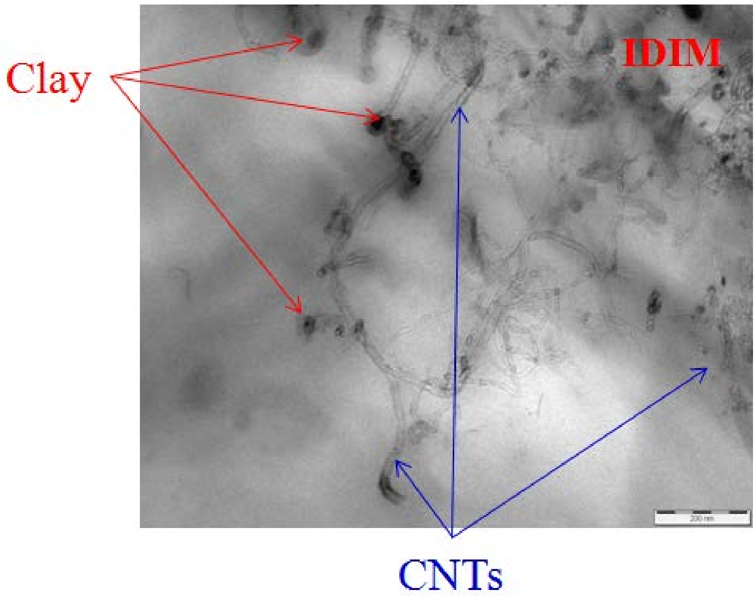


Figure 5.6 TEM image of PP/Clay(4.8wt%)/CNT(0.5wt%) composite

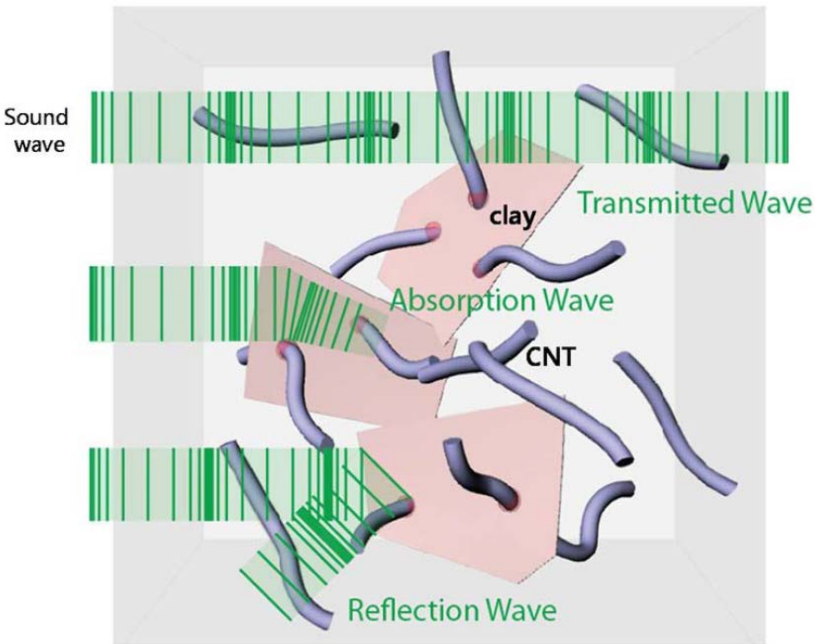
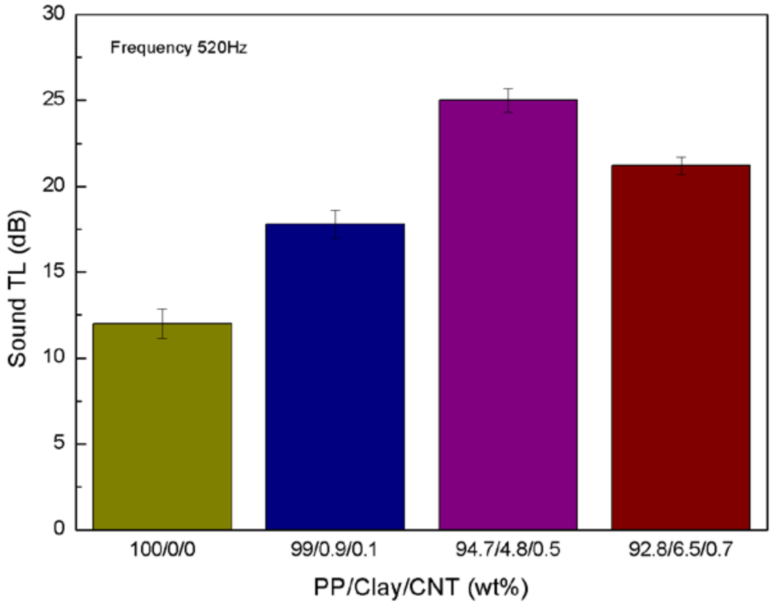


Figure 5.7 Sound wave transmission through PP/Clay/CNT composite [24]

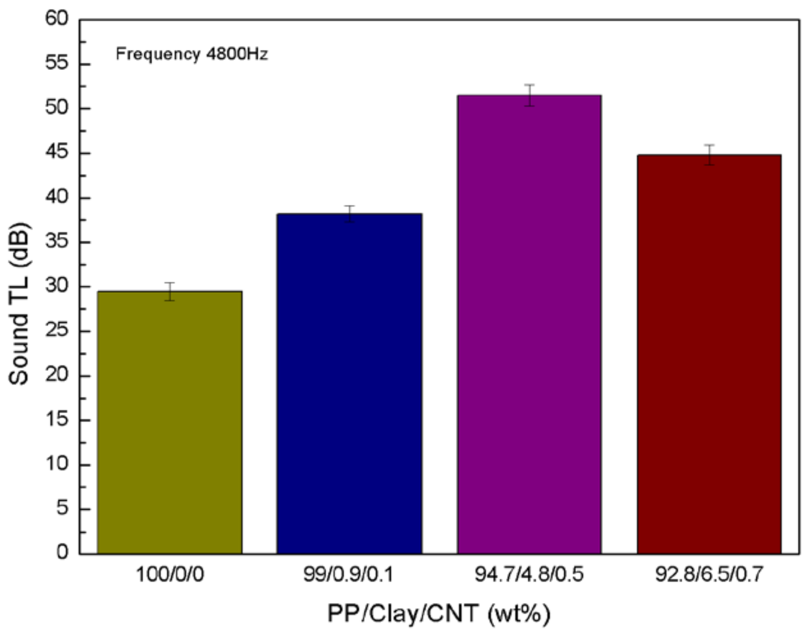
5.3 Synergistic effects as a function of filler concentration

As PP/Clay/CNT nanocomposites showed the highest performance on soundproofing ability, they were selected for further study with regard to the amount of nanofiller added to the polymer matrix.

The sound TL of PP/Clay/CNT composite as a function of filler concentration at 520 *Hz* and 4800 *Hz* is shown in Figure 5.8 (a) and (b), respectively. The magnitude of the soundproofing effect increased with clay and CNT up to 4.8wt% and 0.7wt%, respectively. However, as more loadings of clay and CNT added, the sound TL values began to decrease.



(a) 520 *Hz*



(b) 4800 *Hz*

Figure 5.8 Sound TL of PP/Clay/CNT composite as a function of filler concentration

The synergistic effect as a function of filler concentration could be analyzed through TEM image as shown in Figure 5.9. Figure 5.9 (a) shows that the PP/Clay(0.9wt%)/CNT(0.1wt%) composite possessed only a small amount of nanofiller. Therefore, few synergistic interactions were available, which resulted in relatively low soundproofing ability. Conversely, the PP/Clay(4.8wt%)/CNT(0.5wt%) composite exhibited the highest degree of soundproofing due to the synergies resulting from homogeneous dispersion and strong interactions between the fillers and the matrix, as shown in Figure 5.9 (b). Such structural characteristics are the primary causes of soundproofing ability for nanocomposites. As for the PP/Clay(6.5wt%)/CNT(0.7wt%) composite, shown in Figure 5.9 (c), much lower soundproofing ability was realized because the CNT and clay particles were not uniformly dispersed throughout the PP matrix[24].

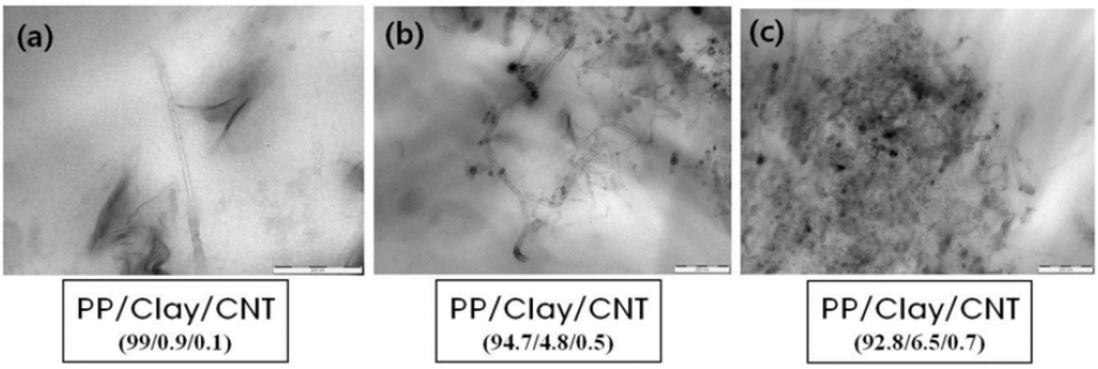


Figure 5.9 TEM image for each weight percentage PP/Clay/CNT composites

Chapter 6 Equation building for soundproofing effect

6.1 Overview

Inorganic particulate-filled polymer composites have good sound absorption and insulation characteristics besides light quality and high specific strength, and so on[80,81]. The sound transmission loss depends on energy loss due to reflection energy and absorption energy from incidence energy. The reflection energy is contributed by the reflections on PP surface and interfaces between PP layers and between clay particles and the PP matrix. The main factors effecting soundproofing property are the density and modulus of pp, the shape and size of clay particles, and the contents between clay particles. More clay particles over a threshold value may move to together and form aggregation which can prevent sound transmission and decrease viscosity. It is necessary to build a mathematical equation including all effecting factors above to reveal the theory or principle for soundproofing property of this kind of nanoclay reinforced PP composites.

6.2 Equation building

The main factors effecting soundproofing property should be embodied as different mathematic parameters for building equations which can show the relationship between sound transmission loss and each effecting factor. The experimental data can be verified through fitting curves drawing according to this equation.

6.2.1 Theoretical hypothesis and parameter determination

In practice engineering application, the sound TL is usually used to describe the sound insulation property of materials. For equation building, the filler particles are considered as ellipse sheet shape with uniform size, and the dispersion of the filler in polymer matrix is uniform [82,83]. The diagram of sound insulation element was shown in Figure 6.1, and the sound wave propagation in PP matrix was shown in Figure 6.2. When sound wave (P_{1i}) projects the surface of PP matrix, a part of it is reflected to the air which is marked as P_{1r}, while the sound wave which enters into PP matrix is called as P_{2t}. When the sound wave spreads through PP matrix, a part of it which maked as P_{2r} will be reflected on the surface of clay particles. And the transmission wave which enters into air again is called as P_t. The sound transmission coefficient ψ can be expressed in Equation (1). As shown in Figure 6.2, from the acoustic boundary conditions at the two special coordinate points X=0 and X=T, the transmission coefficient after sound wave going through PP matrix can be obtained from Equation (2) [83].

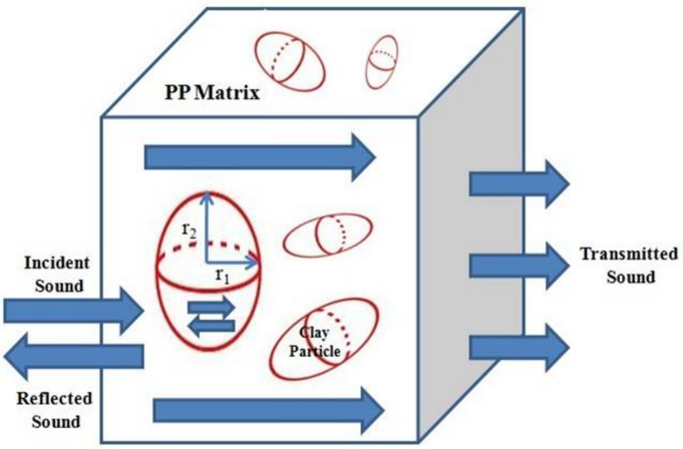


Figure 6.1 Hypothetical modeling with ellipse sheet clay particles in PP matrix [83]

$$\psi = 10\log\frac{1}{\tau_I} \tag{1}$$

$$\tau_I = \frac{4}{4cos^2T+(R_{12}+R_{21})^2 sin^2 T} \tag{2}$$

Table 6.1 shows the meaning of each parameter presented in all equations. The whole sound TL of PP/Clay composites (φ_T) can be divided into two parts: sound TL through PP matrix (φ_P) and clay particles (φ_C). According to the values of characteristic impedance of the air R_0 and combination with equation (1) and (2), the final sound TL of PP matrix can be described in Equation (3). All the values of required parameters are shown in Table 6.2.

$$\varphi_P = 20\log f + 20\log\rho_1 + 20\log T - 42.4 \tag{3}$$

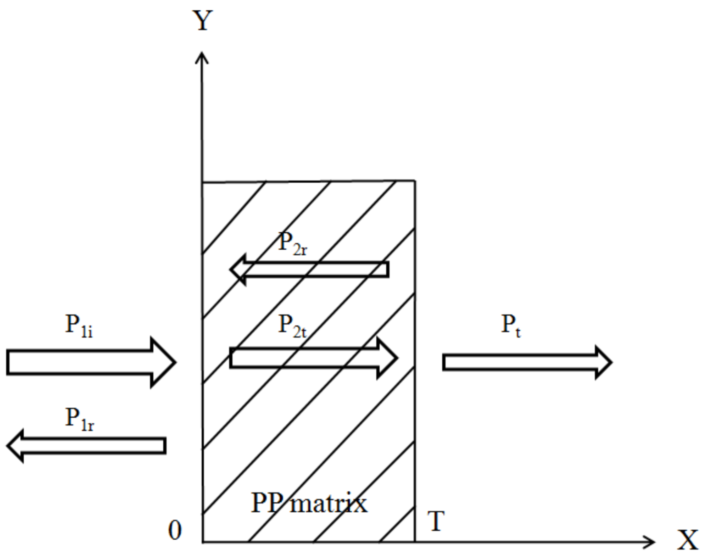


Figure 6.2 Sound wave propagation in PP matrix

Table 6.1 Parameters for equation building

Parameter	Physical Significance
R_{12}	$R_{12} = \dfrac{\rho_2 C_2}{\rho_1 C_1}$
R_{21}	$R_{21} = \dfrac{\rho_1 C_1}{\rho_2 C_2}$
N	$N = \dfrac{R_{21} + R_{12}}{C_2} = \dfrac{E_1\rho_1 + E_2\rho_2}{E_2\sqrt{E_1\rho_1}}$
R_0	$R_0 = \rho_0 C_0$
R	$R = \sqrt{(L/2)^2 + (t/2)^2}$

Table 6.2 Value of related parameters

Parameter		Value							
f (Hz)	Sound frequency	800	1600	2400	3200	4000	4800	5600	6400
v	Weight percentage	0.9wt%			4.8wt%			6.5wt%	
T (mm)	Specimen thickness	3							
L (nm)	Length of clay particle	160							
t (nm)	Thickness of clay particle	8							
ϕ (mm)	Specimen diameter	29							
E_1(Pa)	Young's modulus of PP	1.5×10^9							
E_2(Pa)	Young's modulus of clay	2.0×10^9							
ρ_1 (kg/m^3)	Density of PP	910							
ρ_2 (kg/m^3)	Density of clay	1660							
ρ_0 (kg/m^3)	Density of air	1.2							
C_0 (m/s)	Sound velocity in air	340							

For sound TL through clay particles, the filler particle is considered as taking a medium into a relatively large space, and then the transmission loss through filler particle is calculated. As shown in Figure 6.3, an element cross section is taken from the ellipse sheet, and the sound wave propagation in it is analyzed to derive the transmission loss equation of the element cross section. Here, R is the effective radius of clay particle with ellipse sheet shape. L and t are the length and thickness of clay particle, respectively. The relationship between R, L, and t is described in Equation (4). Therefore, the expression of the transmission coefficient $\nabla_{\tau_{I_1}}$ can be obtained from Equation (5). With integration Equation (5) on the whole cross section, the average transmission coefficient τ_{I_1} can be calculated. And according to Equation (1), the sound TL through clay particles with one element cross section can be obtained from Equation (6)[84,85].

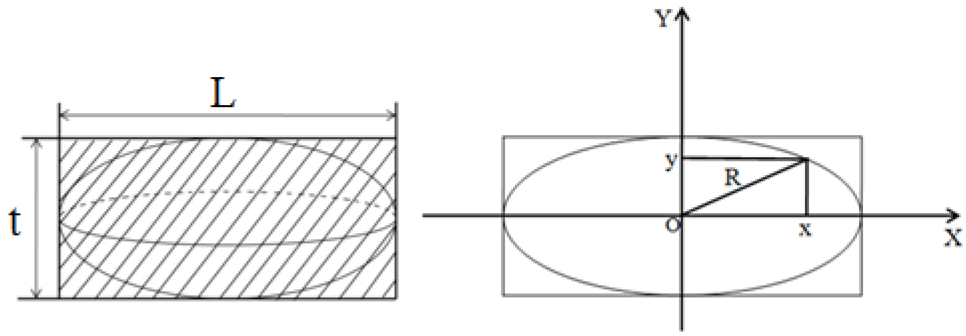


Figure 6.3 Sketch of sound wave transmission in ellipse sheet clay particles

$$R = \sqrt{(T/2)^2 + (t/2)^2} \tag{4}$$

$$\nabla_{\tau_{I_1}} = \frac{1}{1+(R_{21}+R_{12})^2(R^2-x^2-y^2)} \tag{5}$$

$$\varphi_C' = 20\log\left[R(R_{21}+R_{12})\frac{f}{C_2}\right] + 3 - 10\log\left\{\ln\left[(R_{21}+R_{12})^2\frac{f^2}{C_2^2}R^2+1\right]\right\} \tag{6}$$

As for clay particles, the radius is relatively so smaller compared with sound velocity. Therefore, the second part of φ_C' can be ignored. And according the relationship between *C*, *E* and *ρ*, the eqution of sound TL in clay particles can be modified and shown in Equation (7). As the relationship between clay particle volume percentage (V_f)and clay particle weight percentage (v) is described in Equation (8)[85], the whole amount of clay particles (M) in a specimen can be calculated through Equation (9). Therefore, the whole sound TL through clay particles (φ_C) can be described in Equation (10).

$$\varphi_C' = 20\log(RNf) + 3 \tag{7}$$

$$\mathrm{V}_f = \frac{\mathrm{v}/\rho_2}{\mathrm{v}/\rho_2 + (1-\mathrm{v})/\rho_1} \tag{8}$$

$$\mathrm{M} = \mathrm{VV}_f = \pi\left(\frac{\Phi}{2}\right)^2 \mathrm{TV}_f = \frac{\pi\Phi^2\mathrm{TV}_f}{4} \tag{9}$$

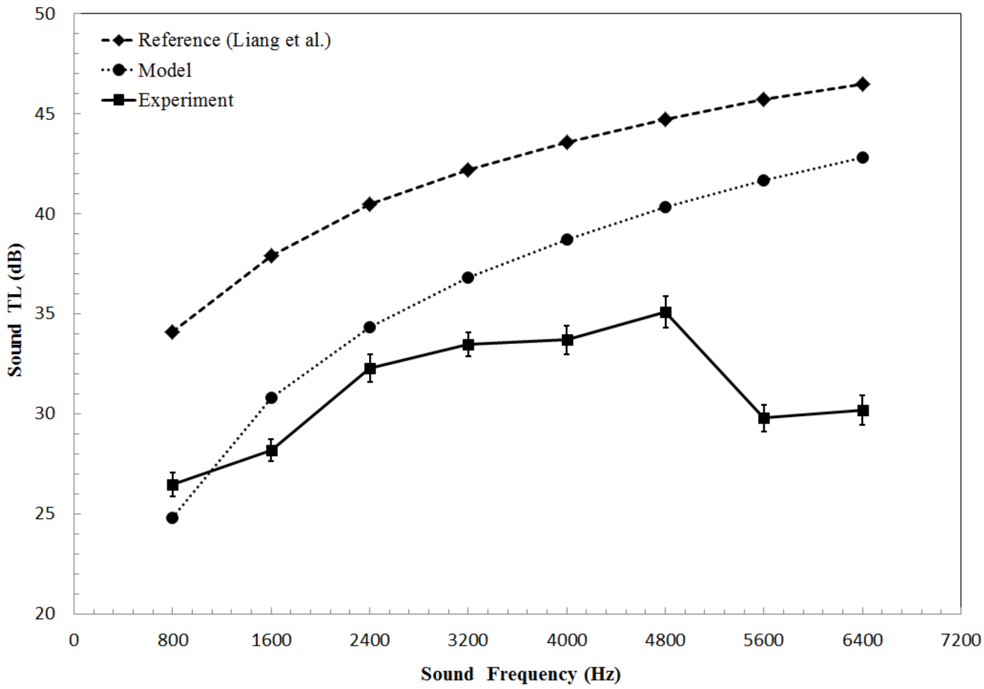
$$\varphi_c = \mathrm{M}\varphi_c' = \frac{\pi\Phi^2\mathrm{TV}_f}{4}|20\log(\mathrm{RN}f) + 3| = \frac{\pi\Phi^2\mathrm{Tv}}{4\rho_2[\mathrm{v}/\rho_2 + (1-\mathrm{v})/\rho_1]}|20\log(\mathrm{RN}f) + 3| \tag{10}$$

Here, sound velocity calculation can be used by elasticity modulus for instead of measurement in polymer matrix and filler particles by equation. The PP molecular chains have alternate deformation due to sound wave transmission effect through PP matrix. Therefore, sound TL can be achieved by partial sound energy changing into heat energy and the whole sound TL of PP/Clay composites calculation process can be described through Equation (11) with variable parameters and modified coefficient due to size unit of clay particles.

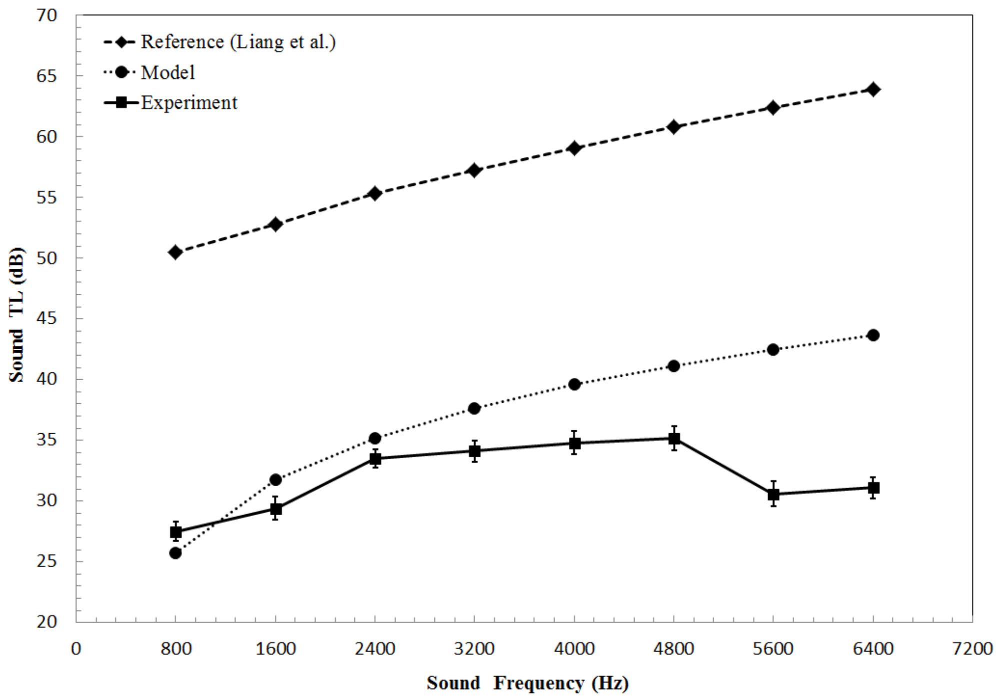
$$\varphi_\mathrm{T} = \varphi_\mathrm{P} + \varphi_\mathrm{C} = 20\big(\log f + \log\rho_1 + \log\mathrm{T}\big) - 42.4 + \frac{\pi\Phi^2\mathrm{Tv}\times10^5}{4\rho_2[\mathrm{v}/\rho_2 + (1-\mathrm{v})/\rho_1]}|20\log(\mathrm{RN}f) + 3| \tag{11}$$

6.2.2 Experiment data verification

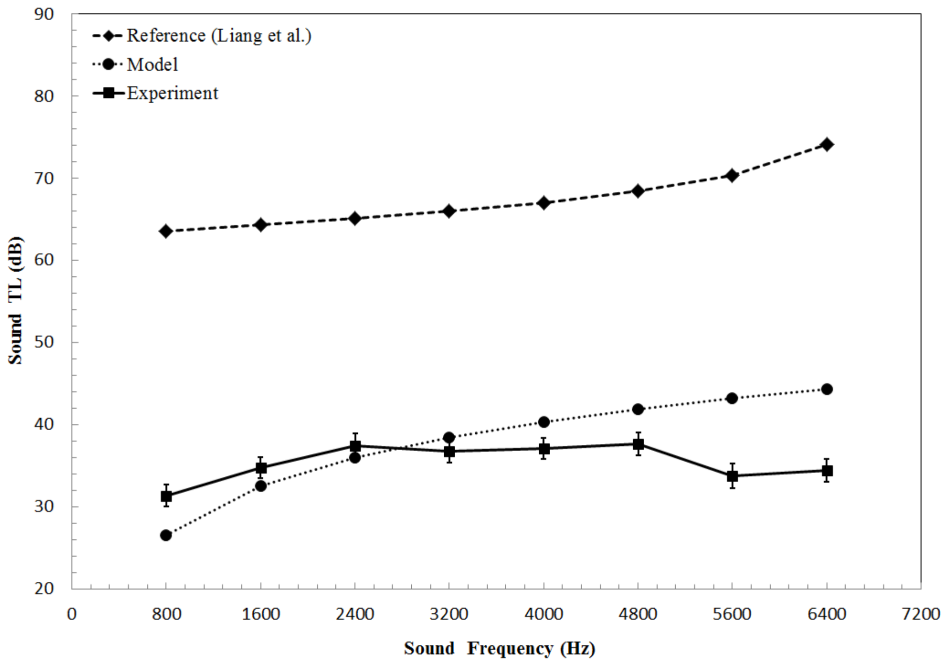
With sound TL experiment at each weight percentage and sound frequency by data fitting method, the final sound TL of PP/Clay composites can be calculated through Equation (11). Figure 6.4 (a), (b), (c) (d), (e), (f) show the comparison of these six group data for diameter 29*mm* specimens. In conclusion, the fitting curve according to modeling equation showed similar trend and perfect fitting values for each weight percentage of clay reinforced PP matrix.



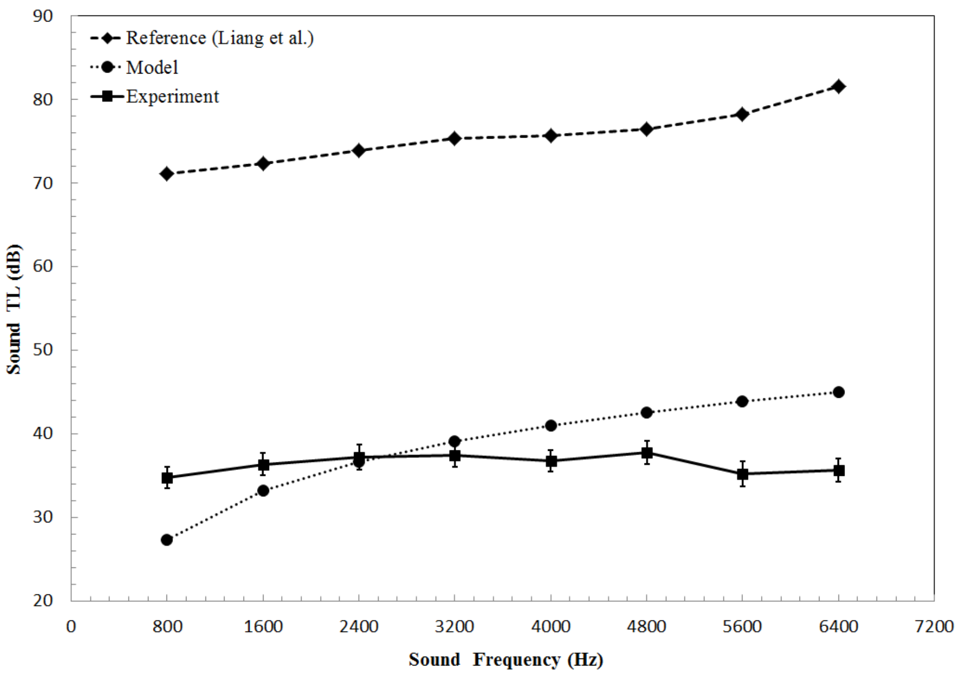
(a) 0.9wt% clay



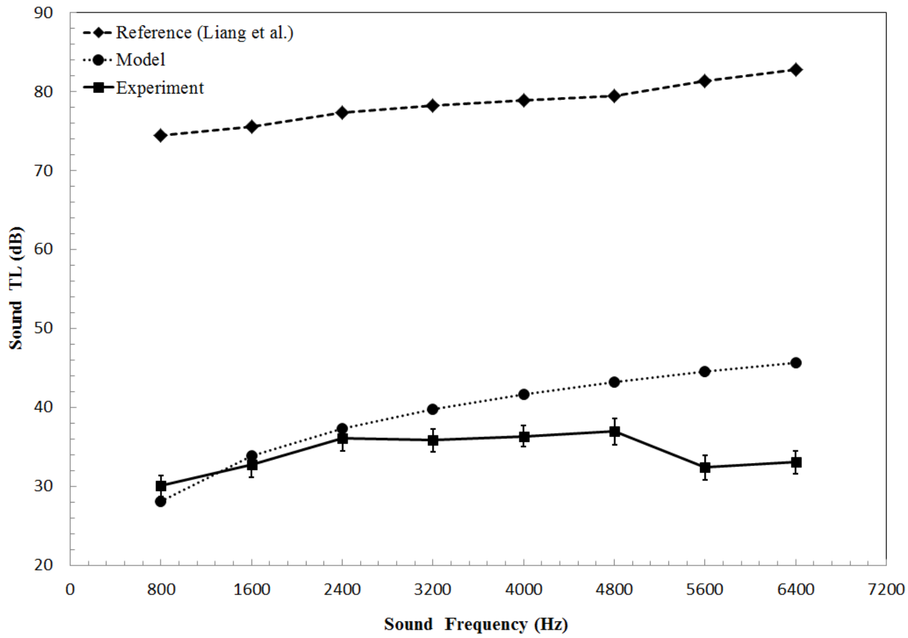
(b) 2.9wt% clay



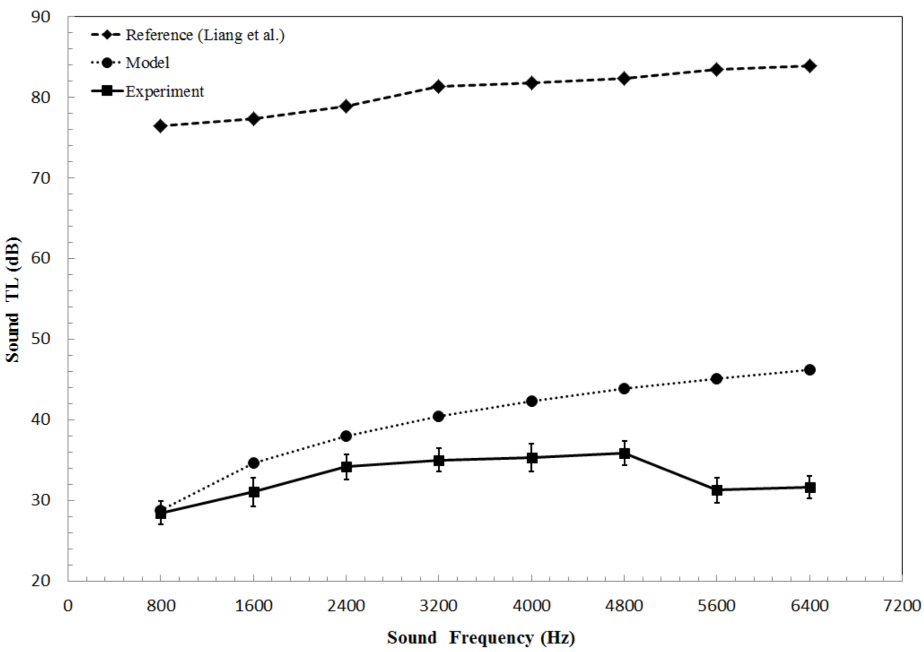
(c) 4.8wt% clay



(d) 6.5wt% clay



(e) 8.2wt% clay



(f) 9.9wt% clay

Figure 6.4 Data comparison for diameter 29*mm* specimens

As seen from Figure 6.4 (a), (b), (c), (d), (e), and (f), the largest error between model data and experiment data is about 41.7%, 40.3%, 28.6%, 26.2%, 37.9%, and 46.1%, respectively. And the largest error between reference data and experiment data is about 75.5%, 105.5%, 114.8%, 128.6%, 150 %, and 168.4%, respectively.

Chapter 7 Applications

7.1 Overview

Recently, a variety of polymer composites have been used in various fields such as automotive industry, aircraft, high-speed train, aircraft, and aerospace engineering increasingly due to their strong mechanical properties and other exceptional abilities such as soundproofing ability. Figure 7.1 shows the use of these polymer composites in various engineering fields. Moreover, soundproofing materials are also widely used in daily life. For example, home appliances like air-conditioner and refrigerator, entertainment places such as recording studio, theatre, piano room, instruments factory, etc.

Figure 7.1 Polymer composites application in various fields

7.2 Soundproofing materials for automotive parts

A variety of polymer composites have been used in various automotive parts. As noise reduction is an important issue in automotive industry, the development and application of various polymer composites with perfect soundproofing property can help solve this issue [86-89]. Figure 7.2 shows the main parts of automotive body which are applied with perfect soundproofing property of polymer materials. As shown in Figure 7.2, the floor plan made of polymer materials with good soundproofing ability can reduce noise caused by uneven roads. For roof, it can reduce noise such as rain tapping. For front fenders and front wheel arcs, it can reduce noise caused by engine, tire, and road gravels. For doors, the soundproofing materials can enhance car audio quality and reduce noise from other passing-by vehicles. Some other parts, such as triangle joint, integral seat, bumper, and transverse support beam, are mostly made of nanofiller-reinforced polymer composites. Among them, PP is the most widely used polymer matrix materials with different nanofiller-reinforcement. These nanofiller-based polymer composites are currently considered as optimal materials for improving the sound insulation performance of automotive components. The interior, chassis, and engine parts of automotive components are commonly manufactured using soundproofing materials. As noise can be produced by engine parts during starting and driving, it is necessary to reduce noise to minimum if possible. Therefore, sound insulating materials are used for fabrication of engine top cover and sound absorbing materials are used for fabrication of engine undercover [90-92].

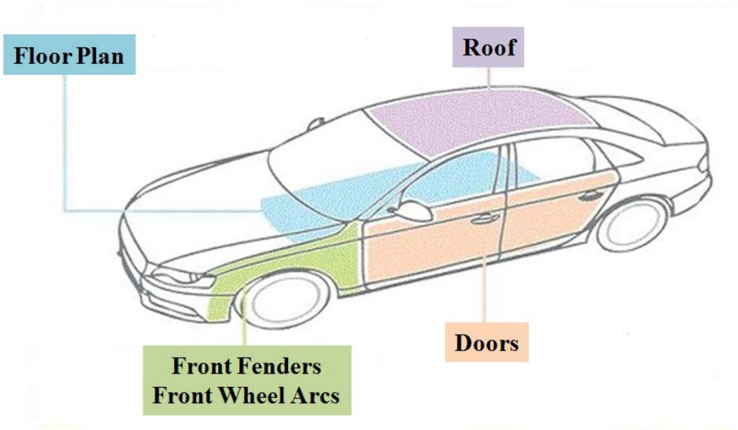


Figure 7.2 Potential application of soundproofing polymer materials on automotive body

7.3 Applications

7.3.1 In automotive

Recently, the need for new composites with the ability both improving mechanical property and reducing noise has arisen much. In this research, the soundproofing and mechanical properties of nanofiller-reinforced PP composite was studied and compared to pure PP. As a result, PP/Clay and PP/Clay/CNT composites were selected as the best specimens with the both good soundproofing ability and mechanical property. In order to compare intuitively, Table 7.1 shows the increasing values for both soundproofing and mechanical abilities of PP/Clay and PP/Clay/CNT composites compared with pure PP. The PP/Clay composite showed suitable mechanical properties and perfect soundproofing ability because of the two-dimensional structures formed between the clay particles and the PP matrix. Furthermore, the PP/Clay/CNT composite showed the highest soundproofing ability due to the synergistic effects of three-dimensional structures formed between the clay, CNT particles and the PP matrix. Anyway, for each kind of nanofiller-reinforced PP composite, the uniform and homogenous dispersion of fillers throughout the PP matrix, and the synergistic effects between the fillers and the PP matrix were the main factors resulting in the improvement of soundproofing and mechanical properties [24].

Table 7.1 Increasing values for nanofiller-reinforced PP composites compared with pure PP

Application Fields	Soundproofing property Composites	Frequency	Increasing values	Mechanical property	Increasing values
Automotive	PP/Clay (6.5wt%)	520Hz	5dB	Modulus	0.7GPa
		4800Hz	10dB	Tensile strenth	5.5MPa
	PP/Clay(4.8wt%) /CNT(0.5wt%)	520Hz	13dB	Modulus	0.5GPa
		4800Hz	20dB	Tensile strenth	6.2MPa

7.3.2 Sound cover for small devices

Due to perfect soundproofing performance of these PP/Clay and PP/Clay/CNT composites, it is possible to make them into package for sound cover using on small devices such as relay and DC cooling blowers as shown in Figure 7.3. The sound frequency range for relay is 2~15 *KHz* and the maximum noise is about 50 *dB*. And the sound frequency range for DC cooling blowers is 200~5000 *Hz* and the maximum noise is about 20~30 *dB*.

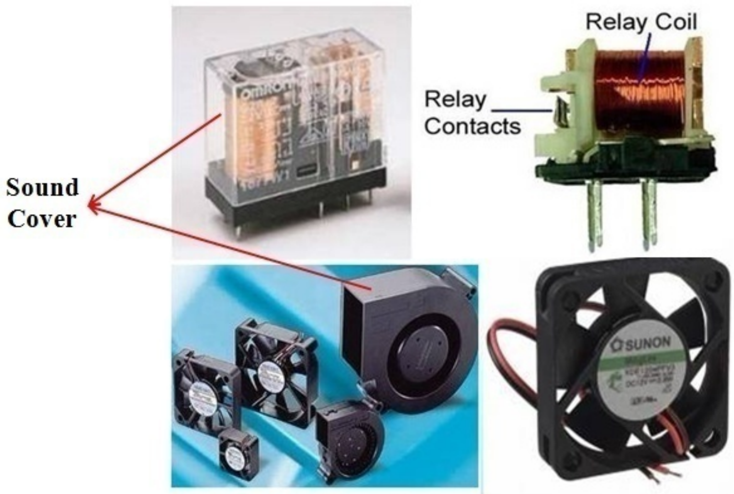


Figure 7.3 Sound cover for small devices

Chapter 8 Conclusions

A series of nanocomposites prepared by mixing various nanofillers into a PP matrix are evaluated in this study for their soundproofing and mechanical properties. These perfect performances result from synergistic interactions between the polymer matrix and the nanofiller particles. Moreover, the preparation of materials, fabrication of test specimens, measurements of soundproofing and mechanical properties, and mathematic equation building with various parameters affecting soundproofing performance are introduced and explained in this thesis in detail.

Solution-blending method was used here to obtain homogenous dispersion of the filler throughout the polymer matrix. The soundproofing properties of PP/Talc, PP/CNT, PP/Clay, and PP/Clay/CNT composites were compared with those of pure PP as a function of filler content and all tested specimens showed better soundproofing effect than pure PP matrix alone. Among all of the test specimens, PP/Clay(4.8wt%)/CNT(0.5wt%) composite yielded maximum sound TL of 15~21 *dB* at 3200~6400 *Hz* due to homogeneous dispersion of clay and CNT particles in PP matrix. As the interaction between PP chains and these nanofiller particles is much stronger than that of pure PP chains alone, it can prevent and block more sound wave transmission and reflection. Therefore, more sound energy will be changed into heat energy during this process and more sound TL will be achieved. In addition, such homogeneous structure also contributed to the dynamic mechanical property such as storage modulus and loss modulus. In general, 6.5wt% clay was the optimal amount for reinforcement of pure PP and the PP/Clay(6.5wt%) composite showed the best soundproofing property among series of PP/Clay composites.

TEM micrographs were provided for internal mechanisms observation of specimens for soundproofing test. The interactions between the fillers and the polymer matrix resulted in perfect soundproofing abilities because of homogeneous dispersion of filler particles and synergistic effects.

A mathematical equation was built for simulation and comparison including relative parameters affecting soundproofing properties such as weight percentage of filler material, thickness of the specimens, radius of filler particles, sound velocity in matrix material and filler material, density of matrix material and filler material, and diameter of the specimens. The fitting curve according to modeling equation showed similar trend and perfect fitting values for each weight percentage of clay reinforced PP matrix.

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초록

일반적으로 소음은개인과 사회 구성원 모두에게 심각한 피해를 주는 불필요한 공해로 인식되고 있다. 그래서 요즘에 방음재료를 이용한 소음 대책은 다양한 공학분야에서 많은 주목을 받고 있다. 최근에는 폴리머(polymer) 흡수재료가 일상 생활의 많은 분야에서 방음재료로 활용되고 있다.특히, 방음효과가 뛰어난 폴리머 흡수재료의 최대 장점은 경량, 고강도 및 저비용의 특성을 가진다는 것이다.

이번 연구에서는 방음과 기계적 물성을 조사하고 이해하기 위해서 클레이(clay)로 성능이 보강된 폴리프로필렌(PP)을 혼합한 다양한 종류의 나노 복합재료를 만들었다. 일반적으로, 클레이는 소량 사용함에도 불구하고 매트릭스 내부에 고루게 분산되어 이를 통해 급격한 기계적 물성 등의 향상을 발생시키는 나노미터 크기의실리케이트고유 특성에 따라 보강용 필러로널리 사용되고 있다.이와 함께, 폴리프로필렌은 저밀도와 고가열변형온도 특성에 따라 폭넓게 매트릭스 재료로 사용되고 있다.

다양한 방음 물성 측정을 위해 사출성형기로 PP/Clay 시편을 가공하여임피던스 튜브 장비를 이용한 음향 전송손실값(TL)을 측정하였다. 특히, 폴리프로필렌 매트릭스 내부에 적정 성능 구현을 위한 최적의 투입량에 대해폭넓게 조사하였다. 이를 위해, 클레이혼합양을 변화시킨 PP, PP/Clay(0.9wt%), PP/Clay(4.8wt%), 및 PP/Clay(6.5wt%) 시편들의 방음 특성에 대해서 고주파에 사용되는 직경 29mm 시편을 이용하여 실험하였다. 그 결과, 방음 성능 효율이 6.5wt% 클레이 물질을 혼합할 때까지 향상되었다. 특히, PP/Clay(6.5wt%) 시편은 PP 시편 대비 29mm 시편을 이용한 3200~6400 Hz 에서 8~20 dB 의 TL 값 증가율을 보였다. 일반적으로, 6.5wt% 클레이 양이 폴리프로필렌 매트릭스에게 최적의 양이며 PP/Clay(6.5wt%) 시편이 방음과 기계적 물성에 가장 좋은 성능을 보인다. 기계적 특성에 의하면 클레이 필러 많을수록 시편의 저장탄성률과 손실탄성률은 더욱 양호한 탄성을 나타낸다.

방음 물성에 영향을 주는 인자를 확인하기 위한 수학식이 주파수 변수와 클레이 양을 통해 개발되었다. 더구나, 이 수학식은 실험값과 유사한 완벽한 곡선 일치를 보인다. 게다가, 이런 PP/Clay 를 이용한 복합재료는 장차 컴퓨터 및 자동차 내부 부품 등에 폭넓게 사용될것이다.

주요어: 폴리프로필렌/클레이, 방음효과, 기계적 물성, 시너지 효과, 수학식

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