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A short review on synthesis and characterisation of nano SiO_2/TiO_2 composite for insulation application

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Abstract

Silica dioxide (SiO_2) and titanium dioxide (TiO_2) are nanoparticle fillers that are widely incorporated into polymer matrix for thermal insulation application. Combination of both fillers in producing polymer nanocomposite is interesting to review. This paper reviews on the current and recent research on the method to incorporate the SiO_2/TiO_2 nanoparticles as the fillers into various polymer matrix such as direct mixing, intercalation, sol-gel and in situ polymerisation as well as the effect of nanofillers on the thermal properties, morphology studies, rheology behaviour, mechanical property, and conductivity (thermal and electrical) of the SiO_2/TiO_2 polymer nanocomposites. This paper also reviews the effect of SiO_2/TiO_2 nanoparticles to the polymer nanocomposites in term of dielectric properties as a potential electrical insulation material. SiO_2 nanoparticles presented to be the best filler to enhance the dielectric properties compared to the TiO_2. When both of nanofillers are incorporated into the polymer matrix, a better result in term of mechanical, thermal, and electrical insulation properties are produced.

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1.0 Introduction

Recently, nanoparticle fillers such as silica dioxide (SiO_2) and titanium dioxide (TiO_2) have gained a great deal of attention. These nanofillers are replacing the conventional fillers such as glass beads, ceramic beads, expanded perlite, sepiolite, and poroesaus silica ceramic due to simpler in application, lower in cost and shows enhancement of thermal insulation properties (Sun et al., 2021). Conventional fillers were reported on particle poor dispersion and particle aggregates which leads to the deterioration of the properties of the polymer composite (Madidi et al., 2018). In contrast to conventional fillers, the nanometer-sized fillers (nanoparticle fillers) is the main advantage which is homogeneously may disturbed within the polymer matrix (Syatirah et al., 2020). SiO_2 is one of the fillers that is commonly used and suitable for various applications due to its natural abundance, low cost, high thermal strength, and surface functionality. Meanwhile, TiO_2 is a well-known filler because it is not harmful, chemically inert, has broadband UV filter characteristics and high refraction index, resistant to corrosion and a low-cost material. Incorporation of SiO_2 or TiO_2 had influenced on the polymer properties, especially on the thermal conductivity for thermal insulation application. Polymer composite with incorporated of SiO_2 or TiO_2 particles might be used in very specific areas such as renovation of historical buildings, where not only focus on thermal insulation, but also antibacterial characteristics. Besides, it is widely used in various industries such as energy, aerospace, chemical and metallurgical which contribute to the high demand of thermal insulation materials with excellent properties and high temperature strength (Yu, 2015). On top of that, addition of SiO_2 and TiO_2 in polymer matrix affect the dielectric properties of the polymer nanocomposite. This inspired researchers to explore SiO_2 and TiO_2 for electrical insulation application.

The review presented in this paper is on the current method on synthesizing nano SiO_2/TiO_2 composite and the effect of the nanoparticles on the thermal properties, morphology studies, rheology behaviour, mechanical property, and conductivity (thermal and electrical) of the polymer nanocomposite. Thus, the dielectric properties of the polymer nanocomposites are subsequently discussed as the potential application as electrical insulation material.

2.0 Synthesis methods of SiO_2/TiO_2 nanocomposite

Production of homogeneous polymer nanocomposites achieved by the good interaction between the nanofillers and the polymer and very dependent on the processing method applied (Amin & Ali, 2015). Thus, the suitable method to gain the desired results in term of properties are selected and chosen by considering the nature behaviour of the materials that been utilised such as the shape, surface chemistry, mechanical resistance and thermal resistance in order to produce the polymer nanocomposite (Mallakpour & Naghdi, 2018). Four major routes have been well developed to incorporate the nanofillers into the pure polymer matrix as well as the characterisation findings approached by previous research that will be reviewed in this paper which are direct mixing, intercalation, sol-gel and in-situ method.

2.1 Direct mixing

Direct mixing method is a method that involves the operation of two classes of technique which are melt blending and solution blending. Melt blending or compounding is a process where the mixing of the nanoparticles and polymer operating at higher glass-transition temperature (T_g) and the melting point (T_m) of the polymer while the solution mixing requires evaporation of solvent for well dispersion of nanofiller into the polymer matrix (Pleşa et al., 2016). The melt processing method which involves the mechanical mixer in the conventional method such as internal mixer and extruders which have the type of single and twin screw extruders (Pleşa et al., 2018). Pleşa et al. (2016) found that the small amount of the nanofillers could affect the morphology of the composites due to the absence of large agglomerations of 5 wt.% silicalite-1 nanoparticles via melt compounding of HDPE/silicalite-1 nanocomposites by internal mixer at 170 °C with a rotor speed of 60 rpm for 3 min which indicating the excellent dispersion of SiO_2 nanoparticle as the filler into the HDPE matrix (Pleşa et al., 2016). It is important to operate and maintain the temperature of the mixing process below the degradation temperature of the fillers and the polymers to avoid the degradation of the chemicals which obeyed by the researcher to operate below 340 °C as for HDPE application. The same rules applied to the mechanism of incorporating the nano-TiO_2/SiO_2-acrylic composite in the study of water resistance of the composites via blending method where Du et al. (2019) specified the condition of mixing at 100 °C for 30 minutes give a good feedback at 5 wt.% of the fillers as the water resistance is improved via utilisation of the method (Du et al., 2019). Both solution and melt blending are alike where they involved the mixing and stirring mechanism but differ in the technique of processing where solution blending commonly applied magnetics stirring or ultrasonic irradiation which is more controllable compared to the melt blending that usually use mechanical approach such as internal mixer, single or twin-screw extruders that quite hard to control the process. However, both offer similar quality of dispersion of nanofillers into the polymer matrix even the condition to process the raw materials are different. Solution blending need a compatible solvent to ensure a well dispersion of fillers unlike the melt blending which just need to melt the polymer to disperse the fillers to form polymer composites.

2.2 Intercalation

Intercalation method is a process which includes the exfoliation of the layered particles where the nanofillers, commonly nanoclays, are immersed into a solvent consists of melted polymer where the solvent moving into the layers of the nanofiller to promote exfoliation before evaporation takes place for the filler to disperse into the polymer matrix (Pleşa et al., 2018), (Pleşa et al., 2016). Previous study by Pleşa et al. (2018) implemented intercalation method in order to create alkyd resins/TiO_2/SiO_2 nanocomposites with the concentration of 2 and 4 wt.% of TiO_2 and SiO_2, respectively, has shown a success method in nanocomposites processing where the well dispersed of the fillers into the polymer matrix formed less of aggregation (Pleşa et al., 2018). As claimed in the study of graphene/polymer composites Pleşa et al. (2018) decided to specified the composition of the graphene as a filler must be limited to 1wt.% in order to disperse homogeneously in the polymer matrix due to the low potential in dissolution of the particles (Pleşa et al., 2018). The method of processing polymer nanocomposite by intercalation method applied by Grozdanov et al. (2019) was set at speed of 400 rpm for 30 min at room temperature by a high speed disperser but a dispersion of the nanofillers has a significance problem to the polymer matrix where dispersant agent needed to solve the issue (Grozdanov et al., 2019). Intercalation method can be viewed as a process that need extra suitable agent to ensure the smooth and uniform dispersion of the TiO_2 and SiO_2 into the polymer matrix. Similar to the solution blending in the direct mixing method, intercalation method needs a

solvent to act as the agent for the fillers and polymers to perform the exfoliation and intercalation where the evaporation of the solvent take place at the end of the process to obtain the desired polymer composites. However, the processes favoured a different type of fillers which some of fillers are not suitable to be processed by intercalation method even suit the blending method. It is known that intercalation method is suitable for graphene filler which contrast to blending method which can use wider type of fillers.

2.3 Sol-gel

Sol-gel is a method requires two chemical reaction which are hydrolysis of the metal alkoxides (sol) and condensation of the hydrolysed intermediates (gel) (Pleşa et al., 2018). This method also favourable for the production of inorganic metal oxides to utilised in the creation of inorganic-organic hybrids (Pleşa et al., 2018).TiO_2-SiO_2 which is one of the photocatalytic composition found to form an anatase thin film by Regalado-Raya et al. (2018) on the methanol oxidation process as it dried for 24 hours in room temperature and calcined for 5 hours at 550 °C with the utilisation of stirring plate and an ultrasonic cleaner (Regalado-Raya et al., 2018). Other study of production of SiO_2/TiO_2 core-shell nanoparticles via sol-gel method where Budiarti et al. (2017) reported that the shell partially cover both of the nanoparticles after stirring process at 10 °C for 6 hours before dried at 100 °C and calcinated at 600 °C for the same period of 6 hours (Budiarti et al., 2017). Venckatesh et al. (2012) stated that the PVA/SiO_2/TiO_2 nanoparticles synthesised by sol-gel method under vigorous stirring for 6 hours and dried at room temperature with pH range of 1 to 2 resulting in the agglomeration of titania-silica particles with an average size of 10 to 15 nm which believed to produce non-uniform morphology behaviour (Venckatesh et al., 2012). The sol-gel method proved to be a possible route to process the SiO_2/TiO_2 polymer nanocomposite involving very high temperature of calcination for 5 to 6 hours to obtain a well dispersion of fillers on the composites. Sol-gel is in common method as the other method reviewed in this study when it comes to the mixing mechanism, but it is opposite in the concept where sol-gel need hydrolysis, polycondensation, gelation, aging, drying and crystallisation which more complicated compared to the other method.

2.4 In-situ polymerisation

In-situ polymerisation is a method which usually bring into service for the creation of nanocomposites focusing on thermosetting polymer such as epoxides (Pleşa et al., 2018). In-situ polymerisation is effectively reacted which can be identified by the curing agent to incorporate, initiator or increasing the temperature (Pleşa et al., 2018). As mentioned in the literature, this process is controlled by the temperature, thermal and the properties of the colloidal sols to obtain the desired size of the nanoparticles based on the thermosetting polymers.

The dispersion behaviour of the nanoparticles composites in previous study using PCL as the polymer matrix and SiO_2–TiO_2 particles to produce PCL/SiO_2–TiO_2 nanocomposites via in-situ method can be referred in (Pleşa et al., 2018). This in-situ method reported large aggregation of particles with the addition of more than 10 wt.% fillers due to the phase separation and homogeneity; but well fine dispersion is recorded at the addition of less than 10 wt.% of fillers (Pleşa et al., 2018). The success dispersion by Wu (2007) at the stated concentration of fillers is by specifying the condition of the process at a rotor speed of 50 rpm and a temperature of 90 to 100 °C (Wu, 2007). Besides, a study of the thermosetting polymer in preparing the composites based on epoxy and SiO_2/TiO_2 nanofillers revealed that those nanofillers with the size of 22 nm are well dispersed with no agglomeration in the resin (Pleşa et al., 2018). The study on the literature of in-situ processing method has shown that the process is very compatible with thermosetting polymers especially epoxy resins and the use of SiO_2/TiO_2 nanoparticles as in the current study. In-situ polymerisation is quite similar to the intercalation method as it involved exfoliation and intercalation, but both are different in the term of polymerisation process where in-situ needs an organic initiator or radiation. It can be seen that all process which are the direct mixing, intercalation, sol-gel, and in-situ polymerisation need the similar application which is mixing process but with different speed condition. The similarity of those processes, however, produced a different result of dispersion behaviour of the nanofillers in the polymer matrix.

Processing method of polymer nanocomposite can be done with various methods such as mentioned in this section due to their specialty and advantage along with their specific condition in operating the method as shown in Table 1. Melt processing in this current research study is suitable for the materials to be operated below the degradation temperature and widely used in the incorporation of polymer

nanocomposites (Mallakpour & Naghdi, 2018). This method is also lower in cost and provides high productivity for the incorporation of thermoplastic polymer compared to the sol-gel method which is more likely applicable for organic fillers and in-situ that are compatible with thermosetting polymers (Chae et al., 2006). The in-situ polymerisation is limited due to the limited availability of monomer and compatible solvent suitable to be utilised in the processing. The most conventional way of incorporating SiO_2/TiO_2 nanoparticles into the polymer matrix is by direct mixing either via melt blending or solution blending. However, many other methods have also been applied such as the ultrasonication (Rafiq et al., 2016; Musa et al., 2013; Viswanathan & Chandrasekar, 2018; Rahman et al., 2019b; Yan et al., 2012; Alapati & Joy Thomas, 2012; Amin et al., 2018).

3.0 Effect of incorporation of fillers on polymer composites

Various possible methods of characterising the SiO_2/TiO_2 polymer nanocomposites as performed by most researchers with a successful desired result were obtained. The characterisation of polymer composites is dependent on the properties of the single chemical used to select the suitable operating condition for some equipment such as melt flow index (MFI), differential scanning calorimetry (DSC), thermal gravimetric analysis (TGA) and electrometer while the most constant parameter used in the various polymer nanocomposites is by Fourier Transform Infra-Red (FTIR) and Field Emission Scanning Electron Microscopy (FESEM). The method of characterisation for the SiO_2/TiO_2 polymer nanocomposites are summarised in the Table 2.

Table 1: Methods to synthesis of polymer/SiO_2/TiO_2 nanocomposites

System	Method	Condition	Advantage	Limitation	References
HDPE/silicalite-1	Direct Mixing	170 °C, 60 rpm, 3 min	Environmental friendly, Solvent-free, Suitable for mass production	Limited applications to polyolefins, Long mixing time	(Chae et al., 2006)(Chae et al., 2006)
TiO_2/SiO_2/Acrylic	Direct Mixing	100 °C, 30 min			(Du et al., 2019)
Alkyd resins/TiO_2/SiO_2	Intercalation	400 rpm for 30 min, room temperature	Homogeneous dispersion of fillers	Consume huge amount of solvent	(Grozdanov et al., 2019)
SiO_2/TiO_2 core-shell	Sol-Gel	10 °C, 6 hours, dried at 100 °C, calcinated at 600 °C	Simple, low temperature, High homogeneity of chemicals	Greater shrinkage, Lower number of voids	(Budiarti et al., 2017; Regalado-Raya et al., 2018)
PVA/SiO_2/TiO_2	Sol-Gel	Vigorous stirring for 6 hours			(Venckatesh et al., 2012)
PCL/SiO2–TiO2	In-Situ Polymerisation	50 rpm, 90 to 100 °C	Easy procedure, Uniform dispersion of fillers	Hard to control the polymerisation, Agglomeration	(Pleşa et al., 2018),

Table 2: Characterisation method of SiO_2/TiO_2 polymer nanocomposites

Characterisation	Method	Condition	References
Rheological Behaviour	Melt Flow Index	125 °C–300 °C, pressure of 0.46 to 30.4 kg/cm^2	(Shenoy & Saini, 1986)
Thermal Properties and Crystallisation	Differential scanning calorimetry (DSC)	170 °C, 5 min	(Awad et al., 2019)
Chemical Bonding	Fourier Transform Infra-Red (FTIR)	Wavelengths 4000–400 cm^{-1}	(Jaroenworaluck et al., 2012)
Morphology	Field Emission Scanning Electron Microscopy (FESEM)	10–15 kV	(Nasir et al., 2015)
Thermal Stability	Thermal Gravimetric Analysis (TGA)	30– 800 °C	(Chae et al., 2006)
Conductivity	Electrometer	30–50 °C	(Kadhim, 2015)

3.1 Rheology behaviour

Rheological behaviour is a study which are related to the distortion or deformation of polymer chain by the utilisation of melt flow index (MFI) tester where it provides basic knowledge of material flow with respect to the unit time and viscosity change due to nanoparticle addition. Mohebbi et al. (2014) conducted a measure of rheological behaviour of SiO_2 and TiO_2 nanoparticles incorporated with polystyrene (PS) polymer at 100 °C with the angular frequency of 10^{-2} to 102 s^{-1} for melt flow behaviour identification. The investigation revealed the incremental of storage modulus G′ as the effect of the increasing in frequency and concentration of nanoparticles (0, 1, and 5 wt.%) where TiO_2 nanoparticles observed to enhance the polymer nanocomposites better than the SiO_2 nanoparticles and emit the similar G′ when the two particles combined in the PS polymer. Different results obtained by Khan (2018) in the oil recovery applications where the 1 wt.% PAM polymer with a filler of SiO_2 and TiO_2 showed an increasing trend of viscosity when the concentration of the fillers increased where the TiO_2 nanoparticles reported to increase the viscosity of polymer higher than the SiO_2 nanoparticles.

Both SiO_2 and TiO_2 nanoparticles as fillers affecting the rheological behaviour of the polymer composites in term of storage modulus and viscosity. It is discovered that the TiO_2 nanoparticles gives big impact to the polymer composite rather than SiO_2 fillers even when they are combined to be in one system of polymer composite (polymer/TiO_2/SiO_2) which means that TiO_2 nanoparticles acts as dominant chemicals. The domination of the particles might be due to the properties it holds as a metal oxide substance.

3.2 Thermal properties

The thermal properties and percentage of crystallisation of the polymer composite are usually characterised by Differential Scanning Calorimetry (DSC) to observe the properties such as the cryogenic glass transition temperature (T_g) and the melting and crystallisation peaks. The TiO_2 and SiO_2 nanoparticles with different concentration to fill the polymer matrix is affecting the thermal properties of the composites such as discovered by Grozdanov et al. (2019) where the T_g and curing reaction temperature (T_r) shifted to lower value when incorporating the particles in the alkyd resin indicated that enhancement of flexibility of the nanocomposites with high content of fillers occurred as shown in Table 3.

Abdul Kaleel et al. (2011) investigated on the thermoplastic polymer incorporated with TiO_2 nanoparticles revealed that the obvious changes of the melting temperature (T_m) and the crystallinity of the composites affected by the several manipulated variables such as the concentration of fillers, reaction time and pressure on operating the DSC. Table 4 concludes that the decrease in T_m is due to the short-chain branch of the polymer while the increase of crystallinity is due to the higher loading of filler into the polymer matrix.

3.3 Chemical bond

The wide analysis of the chemical bonding that exists in the polymer nanocomposites is done by using the application of Fourier Transform Infra-Red (FTIR) where it identifies and characterises various functional groups of the polymer/TiO_2/SiO_2 nanocomposites by

Table 3: Transition temperature (T_g) and curing reaction temperature (T_r) of alkyd resin with TiO_2 and SiO_2 nanoparticles (Grozdanov et al., 2019; Wang, et al., 2020).

System	T_g (°C)	T_r (°C)
Alkyd resin	32.30	121.30
Alkyd/2% SiO_2	24.80	106.20
Alkyd/4% SiO_2	22.90	102.50
Alkyd/2% TiO_2	24.87	112.00
Alkyd/4% TiO_2	14.87	104.00

Table 4: DSC data for the PE/TiO_2 nanocomposites (Abdul Kaleel et al., 2011)

Variables	Mass of TiO_2 (mg)	T_m (°C)	Xc
-	0	136	67
Mass of TiO_2	15	137	62
Reaction time (60 min)	15	137	64
Reaction time (120 min)	15	135	71
Pressure (2 bar)	15	130	71

Table 5: Density of the composites with the cross section (Varnagiris et al., 2017)

System	Density (kg/m^3)	Cross-section of material
Pure EPS	20.0103	-
EPS + SiO_2	20.0106	500 nm
EPS + TiO_2	20.0108	1 μm

using infra-red light. According to Du et al. (2019), the incorporation of SiO_2 and TiO_2 nanoparticles into the acrylic resin revealed that chemical changes occurred rather than a simple coating where the bond of C=O observed at the peak 1650 cm^{-1}, Si–O–Si near 1080 cm^{-1} and Si–O–Ti at 975 cm^{-1}. Nakhaei et al. (2016) found FTIR results that show the chemical bonding created by TiO_2/SiO_2 core-shell nanofibers have the string peak at 550–800 cm^{-1} which contributed by the common adsorption of TiO_2 in anatase phase where indicates the presence of Ti–O–Ti bonds. Si–O–Si bond created at the absorption band of 1095 and 1063 cm^{-1} due to the polymeric vibration of Si–O(Nakhaei et al., 2016). Zhao et al. (2016) has studied the chemical bond by the PI/SiO_2 nanocomposite which revealed that the O–H stretching occurred at the wide peak of 3434 cm^{-1} from the pure SiO_2 nanocomposites. The common stretching vibration of Si–O–Si found at the absorption peak of 1089 and 469 cm^{-1} and C=O stretching of symmetric and asymmetric found in the pure PI at 1711 cm^{-1} and 1777 cm^{-1} (Zhao et al., 2016).

3.4 Morphology study

The morphology study is a method used in the observation of the distribution behaviour of nanoparticles inside the polymer composite by using field emission scanning electron microscope (FESEM). The image of SEM of previous study in measuring the morphology of the SiO_2/TiO_2 film on the expanded polystyrene foam was investigated where Varnigiris et al. (2017) has revealed the mixing of the particles contributing to the difference of thickness which affect a small changes of density of composites as shown in the Table 5.(Varnagiris et al., 2017) Venckatesh et al. (2012) proposed in the processing of polymer composites by using PVA polymer matrix has initiated the agglomeration of the SiO_2/TiO_2 nanoparticles with diameter range of 10 to 15 nm and exhibit a size of 20 to 25 nm for the colloidal PVA/SiO_2/TiO_2 nanocomposites. The SEM images of TiO_2 and SiO_2 nanoparticles effect on the epoxy resin studied by Rahman et al. (2019a) has confirmed the agglomeration of the particles in the size of 100 nm.

3.5 Thermal stability

The identification of the thermal stability of polymer/TiO_2/SiO_2 nanocomposite is usually performed by Thermal Gravimetric Analysis (TGA). TGA under the specified and controlled environment is to measure the change in mass of a sample which requirement of variables such as temperature and time. Du et al. (2019) experimented in production of TiO_2/SiO_2–Acrylic nanocomposite resin in where the result revealed the temperature of 600°C could reduce the mass of composites up to 20% as the result of thermal decomposition at high temperatures (Du et al., 2019). Mohebbi et al. (2014) utilised the TGA analysis to study the effect of the SiO_2 and TiO_2 nanoparticles on the SiO_2/TiO_2/Polystyrene composites indicated that similar effect of single incorporation of filler or combined filler where increment of 50℃ observed in the thermal stability of PS nanocomposite.

TGA analysis used to investigate the thermal stability of polymer nanocomposite is successfully performed in the literature where both fillers which are SiO_2 and TiO_2 nanoparticles loading >10 wt.% are believed to have a less reduction of weight when subjected to high temperature compared to the fillers <10 wt.%. The presented experimental data is brought to an agreement that the thermal decomposition of the polymer nanocomposites can be attained at high temperature compared to the pure polymer which means higher thermal stability of the polymer nanocomposites. It is recommended in the insulating application to achieve higher thermal stability of a polymer nanocomposites as the material to be used in the field.

3.6 Conductivity

Conductivity of materials can be categorised into the thermal conductivity and electrical conductivity. Thermal conductivity is the ability of a material to conduct heat, and it represents the quantity of thermal energy that flows per unit time through a unit area with a temperature gradient of 1° per unit distance. Low thermal conductivity of material is the key for thermal insulation application. Meanwhile, electrical conductivity is a material's ability to carry an electrical current. The electrical conductivity of a polymer composite usually determined and tested by using electrometer where this is important to ensure the materials are compatible to act as insulator. The rule for insulator is possess a low value of conductivity.

3.6.1 Thermal conductivity

Wang et al. (2020) investigated TiO_2 nanoparticle as the filler in epoxy resin polymer matrix. The result shows an increment trend in the thermal conductivity after the higher loading of TiO_2 above 1 wt.%. The drastic decrease of value at the content of 0.1 to 1 wt.%

believed due to the scattering phenomenon of phonons at the interface of the composites (Wang et al., 2020; Kadhim, 2015). Meanwhile, Kim et al. (2016) have compared the thermal conductivity emit by CuNW/ETDS and CuNW@SiO_2/ETDS where the SiO_2 fillers loading with 1 to 2 vol % give positive impact on the value of thermal conductivity. The thermal conductivity is said to be increase at the early coating of the SiO_2 nanoparticles to the polymer composites but then decreased gradually after >2 vol% loads. This outcome is due to the reasons of low thermal conductivity at 1.4 W/m·K of single SiO_2 nanoparticles compared to the CuNW. Thus, it is affecting the thermal conductivity of the whole system of the polymer composites whenever high loading of SiO_2 is introduced (Kim et al., 2016).

3.6.2 Electrical conductivity

Akmal et al. (2013) developed a nanocomposite of LLDPE-NR/SiO_2 which revealed a change in the electrical conductivity as different concentration of nanofillers loading. It is recommended to introduce SiO_2 nanoparticles into the LLDPE matrix at <5 wt.% as it shows good performance as an electrical insulator where the electrical conductivity value decrease (Kim et al., 2016). The loading of fillers >5 wt.% will initiate high electrical conductivity of the polymer composites material thus deteriorate the dielectric properties of LLDPE-NR/SiO_2 nanocomposites (Akmal et al., 2013). However, different result occurred when the polymer is subjected to the <5 wt.% TiO_2 nanoparticles where the electrical conductivity increased. This phenomenon is generally by the properties of TiO_2 itself as a metal oxide and poor dielectric properties which is less compatible as electrical insulators (Akmal et al., 2013). Bi et al. (2016) reported the same trend in incorporating SiO_2 into polymer matrix in production of SiC/SiO_2–W/PVDF where reduction in electrical conductivity from 4.2×10^{-5} to 2.8×10^{-7} S/m (at 100 Hz) found when increasing the concentration of SiO_2 filler. This come to an agreement with the previous researcher where SiO_2 nanoparticles is effective in enhancing the dielectric properties of polymer composite make it suitable as an electrical insulator (Bi et al., 2016).

3.7 Mechanical properties

It is known that a material needs to have good mechanical properties to act as insulator. Amin et al. (2015) has confirmed in his study of SiO_2 that the effect of the filler in nano and micro scale have positive impact on the strength of the epoxy composites in term of tensile, elongation and hardness as shown in the Table 6. The big improvement of the tensile strength and hardness are contributed by 5 wt.% of nano SiO_2 filler compared to the 75% higher concentration of micro SiO_2 filler. The results obvious in the effect of filler scale to promote the best mechanical strength as presented by the nano SiO_2 particles where it reduce the polymer chains activity thus improved the hardness of the materials (Amin et al., 2018; Zeng et al., 2015).

The SiO_2 nanoparticles increased the tensile strength when incorporated into XLPE depending on the amount of filler introduced into the polymer matrix which affecting the mechanical strength due to the dispersion behaviour (Said et al., 2019; Zeng et al., 2015). Table 7 shows the SiO_2/XLPE nanocomposite emit the higher tensile strength compared to the pure polymer where 2.5% of SiO_2 nanoparticle influencing in the highest value of tensile strength. Epoxy/TiO_2-SiO_2 nanocomposites prepared by Takari et al. (2021) using direct mixing method presented the best mechanical properties of the composites at 14.64 wt.% of SiO_2. The properties such as elongation at break, yield strength, ultimate strength and elastic modulus of the nanocomposites found to be 29.3 MPa, 17.72 MPa, 17.43 MPa and 4.34%, respectively. Loading of the filler more than the stated amount influenced in the lower mechanical strength as it forms aggregation and agglomeration on the polymer matrix. A developed PPS-TiO_2@SiO_2 nanocomposite with different size of fillers 25, 60, and 100 nm was studied to compare its breaking strength. The introduction of TiO_2@SiO_2–25 nm into the PPS matrix successfully enhanced the breaking strength at 4.07 cN/dtex compared to the neat PPS at 2.42 cN/dtex. The break strength of the polymer nanocomposites is improved when less than 60 nm size of filler is loaded. Beyond this value, the reverse effect of the low efficiency of PPS spinning will occur. Thus, the SiO_2 nanoparticles as filler in polymer composite seems to has the potential in insulating application as it enhances the mechanical properties of the material (Hu et al., 2015).

Grozdanov et al. (2019) in the previous study of alkyd polymer with TiO_2 and SiO_2 nanofillers dealt with the same phenomenon as others when the roughness, hardness and Young's modulus of the composites are depended on the amount of fillers introduced. As tabulated in Table 8, it is expected that the mechanical properties mentioned is enhanced when the SiO_2 is load less than 4 wt.% while opposite effect

of TiO_2 filler which enhanced the mechanical properties when the amount of filler more than 4 wt.% (Grozdanov et al., 2019). Bi et al. (2016) agreed in the effect of SiO_2 nanoparticles to the mechanical properties of the polymer composite developed which is SiC/SiO_2/PVDF nanocomposites where the tensile strength and Young's modulus of the composite is enhanced from 27.7 to 30.1 MPa and 1.67 to 1.93 GPa (Bi et al., 2016). The mechanical properties of SiO_2/TiO_2 polymer nanocomposite can be enhanced by both fillers with the appropriate concentration of each filler where can be seen and interpreted from recent papers that SiO_2 fillers always performed a better result when it is below the loading of TiO_2 concentration and vice versa (wt.% SiO_2 < wt.% TiO_2).

3.8 Electrical insulation properties

The electrical insulation properties of the polymer composites in this review paper are based on the breakdown strength and the resistivity of the polymer composites. A good dielectric material is favoured at high breakdown strength and resistivity.

3.8.1 Breakdown strength

Breakdown strength is important to determine whether the polymer composites is suitable as insulator or the other way around as it provide the information of electrical potential value at the point of where the material failed to insulate. Rafiq et al. (2016) stated that the incorporation of nano SiO_2 into the mineral oil at 20 vol% by ultrasonic method enhanced the breakdown strength up to 16% (Rafiq et al., 2016). The same field study in nanofluid by Ge et al. (2015) and Viswanathan & Chandrasekar (2018) where the SiO_2 and TiO_2 nanoparticles reported to have good properties in enhancing the breakdown voltage up to 8.2% and 10.4% compared to the micro scale of fillers. This phenomenon is described clearly as in previous research where the sizes of the filler affect its performance in a composites where nano scale filler works better due to the well dispersion on composite (Ge et al., 2015, Viswanathan & Chandrasekar, 2018).

Wang & Li (2019) recommend a concentration of less than 2 wt.% TiO_2 nanoparticles when incorporated into LDPE matrix in order to improve the polymer breakdown strength which reported increment of 16% from 223.28 to 265.55 kV (Wang & Li, 2019). Mohamed Ghouse et al. (2013) experimented on the breakdown strength of neat epoxy resin, epoxy/TiO_2 and epoxy/SiO_2/TiO_2. The result was found that the epoxy resin without fillers having lower breakdown strength value compared to the epoxy composites with fillers especially when introduced to both SiO_2 and TiO_2 particles as it. Thus, the epoxy/SiO_2/TiO_2 nanocomposites is agreed to provide dielectric properties when it value enhanced up to 106.2 kV higher than the neat epoxy at 89.2 kV (Mohamed Ghouse et al., 2013). The SiO_2 and TiO_2 nanoparticles could improve the dielectric strength of the zepoxy resin nanocomposite when subjected to the 0.5 wt.% of combined filler (Rahman et al., 2019b). However, the same amount of loading filler conducted by Abdel-Gawad et al. (2017) reacted opposite to the breakdown strength of PVC polymer composite when only TiO_2 nanoparticles introduced into the matrix but improved at above 5 wt.%. The same polymer composite investigated by Habashy et al. (2019) where

Table 6: Mechanical properties of neat epoxy, micro and nano epoxy composites (Amin et al., 2015)

System	Tensile strength (MPa)	Elongation (%)	Hardness (%)
Neat epoxy (NEP)	64.2	110 %	54%
20%SiO_2/Epoxy Micro-composite (EPMC)	65.7	Decrease of 8%	Increase of 3.7%
5%SiO_2/Epoxy Nanocomposite (EPNC)	69.3	Decrease of 12%	Increase of 9.2%

Table 7: Tensile strength of neat XLPE and XLPE/SiO2 nanocomposites (Said et al., 2019)

System	Tensile strength (N/mm^2)
Neat XLPE	15
2.5%SiO_2/XLPE	19
4%SiO_2/XLPE	18

Table 8: Mechanical properties of pure alkyd, alkyd/TiO_2 and alkyd/SiO_2 nanocomposites (Grozdanov et al., 2019)

System	Roughness (nm)	Hardness (MPa)	Young's Modulus (GPa)
Alkyd resin	54	51	1.681
Alkyd/2% SiO_2	346	52	1.957
Alkyd/4% SiO_2	344	39	1.464
Alkyd/2% TiO_2	98	57	2.011
Alkyd/4% TiO_2	410	45	2.179

PVC composite is enhanced it insulating properties in term of breakdown strength up to 7% by introducing the 5 wt.% of SiO_2 nanoparticles (Habashy, et al., 2019). Yan et al. (2013) discovered the potential of SiO_2 nanoparticles incorporated in epoxy resin as insulation properties when the breakdown strength of the composites increase up to 27% compared to the pure epoxy resin(Yan et al., 2013). The same experiment did by same author when incorporated 3 wt.% of nano silica into epoxy resin matrix which improved the breakdown strength at high value from 52 to 116 kV/mm (Yan et al., 2012). The favourable polymer used in dielectric study which is epoxy polymer studied by Awais et al. (2019) discovered an increase in dielectric strength of epoxy/TiO_2/SiO_2 nanocomposite when less than 0.6% filler was used which emit highest value at 395 kV/mm (Awais et al., 2019). Some of the investigation used different scale of fillers to compare the breakdown strength of the polymer composites which gives significance impact to the value as studied by Amin et al. (2018) which reported the increment of 8% and 19% of the neat epoxy breakdown strength when load micro and nano SiO_2 particles, respectively (Amin et al., 2018).

In the study of PP/EDM composites, Hidayah et al. (2015) found that the dielectric breakdown strength is majorly influenced by the SiO_2 followed by TiO_2. This might be due to the properties of SiO_2 that suit the insulators application compared to the metal oxides (Hidayah et al., 2015). Mohan et al. (2019) optimised the condition of filler into polymer matrix to produce an improved dielectric breakdown strength at 84.76 kV/mm by the load of 3 wt.% of TiO_2 nanoparticles (Mohan et al., 2019). Increase in the filler concentration will lead to agglomeration of nanoparticles which then decreasing the dielectric breakdown strength (Polizos et al., 2010). Mathew et al. (2018) experiment in PMMA/TiO_2 nanocomposites gives the view of the potential insulation properties of the materials as the time for the breakdown to occur for more than 4 minutes at above 120 °C. The increase in filler content contributed to the low breakdown strength which tells the composites to have high relative permittivity. This could affect the materials to be applied in the insulation field as the properties for insulation is low in relative permittivity (Mathew et al., 2018). Breakdown strength of PVC/TiO_2 nanocomposite enhanced up to 11.6% from the pure PVC with the loading of 0.5 wt.% TiO_2 nanoparticles confirmed by Abdel-Gawad et al. (2020).

3.8.2 Resistivity

Ge et al. (2015) confirmed that SiO_2 and TiO_2 nanofillers increased the resistivity of pure grease which performed as a good insulator in the nano scale of fillers compared to the micro scales as (Ge et al., 2015). In term of resistivity, Viswanathan & Chandrasekar (2018) found SiO_2 influenced more at 0.01 wt.% to the volume resistivity of the mineral oil system. (Viswanathan & Chandrasekar, 2018)Rahman et al. (2019b) discovered the insulation properties of the SiO_2/zepoxy nanocomposites to improve at less than 0.8 wt.% with the highest value recorded at 0.6 wt.% was 3.4×1014 Ω·cm. The same test used for TiO_2/zepoxy nanocomposite where it emits lower resistivity when subjected to high amount due to the nature of titanium as a conductive material but still can have the maximum resistivity at 0.4 wt.% TiO_2 nanoparticle. When both of the particles combined into one system of nanocomposite, the positive trend of resistivity appeared which 0.8 wt.% of fillers improved the resistance up to 23% and this could be a good material to be applied in insulation field (Rahman et al., 2019b).

Awais et al. (2019) indicated that the SiO_2 and TiO_2 nanofillers performed better in study of dielectric properties when combined with the polymer matrix. The resistivity increased proportionally to the amount of fillers introduced where the maximum concentration at 0.8 wt.% enhanced the resistivity up to 24% from 2.9×10^{16} Ω·cm without filler to 3.7×10^{16} Ω·cm (Awais et al., 2019). Wu et al. (2017) discovered that the SiO_2 nanoparticles does give the biggest impact to the polymer composites to act as insulator. Zeng et al. (2015) developed the insulator materials from MWCNT@SiO_2 at 0.8 wt.% which resulting in resistivity of 2.36×10^9 Ω·cm indicating that it appears as an insulator which rules to be more than 1.0×10^9 Ω·cm. Meanwhile, Said et al. (2019) discovered the XLPE/SiO_2 nanocomposites to have high volume resistivity at 2.5 wt.% of SiO_2 nanoparticle.

The effects of SiO_2 and TiO_2 nanoparticles to the rheology, thermal, chemical bonding, morphology, thermal stability, conductivity, mechanical and insulation properties have been discussed and reviewed where both particles emit different and variety of performance towards the polymer composites. It is found that each of the fillers has its own specialty in enhancing a material for insulation and depending on their concentration and properties carried.

The SiO_2 nanoparticles seems to have a better effect to the dielectric properties of polymer composite as it naturally lows in the conductivity and have a good mechanical strength make it suitable and feasible to be reinforced along with polymer matrix. TiO_2 nanoparticles which the kind of metal oxide naturally carry a bit high of conductivity value and turns out to emit high relative permittivity which is not recommended as an insulator even it is discovered as a material with good mechanical properties. However, the combined SiO_2/TiO_2 nanoparticles which is the focused on this review study found to be high potential in the electrical insulation application. Thus, the reviewed properties of this combined SiO_2/TiO_2 nanoparticles with a polymer matrix shows it suitable to be used as electrical insulator material compared to the single particle used. The observation on the effect of sizes of fillers to the properties of polymer nanocomposites also give a good guidance in the chosen of appropriate scale of particles to produce maximum possible value related to the insulation properties.

4.0 Conclusions

The review of the research trends in SiO_2/TiO_2 nanocomposite by direct mixing, sol-gel, intercalation and in situ polymerisation along with the characterisation findings are presented. In each section, the development of polymer composites based on SiO_2/TiO_2 nanoparticles as the fillers in term of its effect on the properties mentioned such as rheology, thermal, chemical bonding, morphology, thermal stability, conductivity (thermal and electrical), mechanical and electrical insulation properties have been discussed. It is conventional to use direct mixing as the method to incorporate SiO_2/TiO_2 nanoparticles into polymer matrix as it provides better dispersion of fillers as well as the low cost of the process, but the selected method is not limited to direct mixing as different polymer suit different processing method such as sol-gel which is more likely applicable for organic fillers and in-situ that compatible with thermosetting polymers. In term of the effects of the SiO_2 and TiO_2 nanoparticles to the polymer matrix in the insulation application, SiO_2 nanoparticles presented to be the best filler to enhance the dielectric properties compared to the TiO_2 but gives a better result in term of mechanical, thermal, and electrical insulation properties when both of nanofillers combined into the polymer matrix thus offer better insulation materials.

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