



A REVIEW ON SILICONE RUBBER/ MONTMORILLONITE NANOCOMPOSITES

Zoalfokkar Kareem Alobad ¹, Salih Abbas Habeeb ², Muayad Abdulhasan Albozahid ³

thoalfegarkerim@gmail.com, salihabbas61@yahoo.com, muayad.albozadhid@gmail.com

^{1,2} Department of Polymers Engineering and Petrochemical Industries, Faculty of Materials Engineering, University of Babylon, Iraq

³ Department of Materials Engineering, Faculty of Engineering, University of Kufa, Iraq

ABSTRACT

Silicone rubber (SIR) is superior to natural rubbers due to having special features like high chemical stability, heat, abrasion and ozone resistance. Global markets show huge attention to produce and use silicone rubber in many applications such as aerospace, automobile, construction, electric insulators and medical industries. Although silicone rubber is hugely utilized in high voltage insulators, it shows low properties such as mechanical properties. Therefore, fillers are added to silicone rubber in order to reducing cost and improving the surface hydrophobicity, thermal and/or electrical conductivity, thermal and/or electric insulation and mechanical properties. This paper reviews on structure, types, general properties, manufacturing methods, applications of SIR, fillers and silicone rubber nanocomposites. It is focused on the popular micro/nanoparticles for dielectric applications. Organic montmorillonite (OMMT) represents one of the most important fillers that are used for dielectric applications. OMMT is added to SIR to improve the cost and electric and thermal insulation and mechanical properties. Tensile properties of SIR/OMMT nanocomposites increased with increasing the content of OMMT. SIR/OMMT nanocomposites with smaller OMMT size showed the highest compression strength and the lowest tear strength. SIR/OMMT Nanocomposites with high content of OMMT displayed greater thermal stabilities, flame retardant properties, dielectric permittivity and dielectric loss.

Keywords: Silicone Rubber, Montmorillonite, SIR/MMT Nanocomposites, Mechanical Properties, Thermal Properties and Dielectric Properties.

مراجعة على المواد المترابطة النانوية ذات اساس من السيليكون مقواة باطيان المونت موريلونيت

مؤيد عبد الحسن البوزاهد

صالح عباس حبيب

ذوالفقار كريم ال عبيد

الخلاصة

يتفوق مطاط السيليكون على المطاط الطبيعي بسبب ميزات خاصة مثل المقاومة العالية للحرارة والاستقرار الكيميائي ومقاومة التآكل ومقاومة الأوزون. تظهر الأسواق العالمية اهتماما كبيرا لإنتاج واستخدام مطاط سيليكون في العديد من التطبيقات مثل الطيران والسيارات والبناء والعوازل الكهربائية والصناعات الطبية. على الرغم من أن مطاط السيليكون

يستخدم بشكل كبير في العوازل عالية الجهد , فإنه يظهر خصائص منخفضة مثل الخصائص الميكانيكية. لذلك تضاف الحشوات لمطاط السيليكون من أجل تقليل التكلفة وتحسين مقاومة الماء السطحية ، الموصلية الحرارية و / أو الكهربائية ، العزل الحراري و / أو الكهربائي والخواص الميكانيكية. تستعرض هذه المراجعة في التركيب ، والأنواع ، والخصائص العامة ، وطرق التصنيع ، وتطبيقات مطاط السيليكون ، والحشوات ومترابكات السيليكون النانوية. وهو يركز على الدقائق المايكروية او النانوية الشائعة للتطبيقات العازلة. تمثل مونت موريلونيت العضوية (OMMT) واحدة من أهم الحشوات المستخدمة في تطبيقات العزل الكهربائي. يضاف المونت موريلونيت إلى مطاط السيليكون لتحسين التكلفة والعزل الكهربائي والحراري والخواص الميكانيكية. زادت خصائص الشد لمترابكات السيليكون النانوية مع زيادة محتوى OMMT. كما وأظهرت مركبات السيليكون النانوية ذات حجم OMMT أقل قوة ضغط وأقل قوة تمزق. في حين أظهرت مترابكات السيليكون النانوية ذات المحتوى العالي من OMMT تحسن في الثبات الحراري وخصائص مثبتات اللهب والسماحية العازلة وفقدان العزل الكهربائي.

الكلمات المفتاحية: مطاط السيليكون, المونت موريلونيت, مواد مترابكة نانوية ذات اساس من السيليكون مقواة المونت موريلونيت, خواص ميكانيكية, خواص حرارية وخواص عزل كهربائي.

INTRODUCTION

Conventional filled polymers are composed of two or more components, including micrometer-sized fillers that are dispersed within polymer matrix. While, polymer nanocomposites are consisted of nanometer-sized fillers (range of 1-100 nm) dispersion within polymer matrix. Polymer nanocomposites have been vastly used and become more interesting from both researchers and industries due to nano-fillers provide significant property enhancements with very low contents compared with micro-fillers which require greater contents to achieve the same performance Simon et al. (2008). Therefore, there are many applications for polymer nanocomposites due to their unique properties that can be used to achieve high-performance products. Abdulhasan (2018). Silicone rubber nanocomposites can be easy to process, and they have good properties such as thermal and oxidation stability, dielectric properties and resistance to ozone and UV radiation. They are used in many application, including automotive, electronics and aerospace industries Gharavi et al. (2010).

In this review, it was displayed the properties and the types of silicone rubber and the fillers such as montmorillonite nanoclays that were used to reinforce SIR. The aim of this article is to give the reader a scientific overview about silicone rubber/montmorillonite nanocomposites and focusing on the mechanical, thermal and dielectric properties of silicone rubber/montmorillonite nanocomposites.

SILICONE RUBBER

Silicone rubber (SIR) represents an elastomer consisted of silicone together with carbon, hydrogen, and oxygen. Polydialkylsiloxanes considers the basic polymer for silicone rubber (Figure (1)), as such, R groups can be methyl, phenyl, vinyl or trifluoropropyl Shit and Shah (2013). Polydimethylsiloxane (PDMS) represents the most common silicones, this can be attributed to the hydrocarbon methyl groups giving the PDMS hydrophobic and water resistance properties (Figure (2)) SH Kim and EA Cherney (1992), JP Reynders and IR Jandrell (1999). Silicone rubber has ability to retrieve its hydrophobicity when a pollution layer is formed on its surface JongsooKim et al. (2000), Farzaneh (2011). It can attribute the property of hydrophobic recovery of the silicone rubber serves to transmit a low molecular weight siloxane from bulk to surface, and this layer is adsorbed (physically or chemically) on the contaminant Homma et al. (1999). Contaminant particles are encapsulated by low molecular weight siloxane layer which improves moisture absorption resistance of contaminant particles (Figure(3)). Nevertheless, insulators made from silicone rubber have a big problem when they are used in humid atmospheric. This is because wet atmospheric conditions causing a significant reduction

in the properties of insulator surfaces such as hydrophobicity, tracking and erosion resistance. Many researchers Chang and Gorur (1994), Hillborg and Gedde (1999), Zhu et al. (2006), Mohammad Amin et al. (2007) have investigated the effects of the loss and recovery of hydrophobicity property of the silicone rubber on the surface properties of insulators. They found that there are three main factors affect the loss of hydrophobicity property of insulators that are made from polymers. These factors include electrical discharge, adsorption of pollution layers and UV radiation. The most common kinds of silicone rubber which are used for outdoor insulators are room temperature vulcanizing (RTV) and high temperature vulcanizing (HTV). RTV silicone rubber is widely utilized as sizing layer to enhance the contamination performance on the ceramic surfaces depending on hydrophobicity phenomenon. As such, HTV silicone rubber is vastly utilized as a weather shed Chang and Gorur (1994). The curing process of HTV silicone rubber is carried out at high temperature under pressure and with catalyst (peroxide-induced free radicals). While, the curing process of RTV silicone rubber is carried out at room temperature with condensation reaction Muhammad Amin et al. (2007). Both RTV and HTV silicone rubber have low molecular weight polydimethylsiloxane in their structures. 5 wt.% of cyclic low molecular weight polydimethylsiloxane is used to produce RTV silicone rubber. In contrast, 3 wt.% of cyclic and linear low molecular weight polydimethylsiloxane is used to produce HTV silicone rubber. The difference in the properties between RTV and HTV silicone rubber is attributed to form of the low molecular weight polydimethylsiloxane, where, linear low molecular weight polydimethylsiloxane has better diffusion in HTV silicone rubber than cyclic low molecular weight polydimethylsiloxane in RTV silicone rubber Farzaneh (2011). Outdoor insulators made from porcelain and glass are coated with RTV silicone rubbers. This can be attributed to insulators with RTV silicone rubber coating have better anti-pollution properties Cherney (1995). Outdoor insulators can be coated by RTV silicone rubber using different methods such as dipping, painting, and/or spraying. Nonetheless, the main drawback of silicone rubber is a tendency to aging. There are many factors which accelerate the aging process in the silicone rubber including environmental factors (UV, acid rain, temperature and pollution) and electrical factors (electric field and leakage current) Farzaneh (2011).

FILLERS

Tracking and erosion resistance of SIR are good, however, there are several features of SIR can be promoted. It can add particles into silicone rubber in order to minimize the cost and enhance particular properties. With increasing the filler content, the tracking performance increases and hydrophobic properties decreases Cherney (1995), SeokSong and RyouYoun (2005). There are several parameters affect the properties of polymer nanocomposites such as concentration and particle size of fillers, dispersion and orientation of filler in the polymer matrix, interfacial adhesion between polymer and fillers and morphology of the prepared composites materials Deng et al. (1993), SeokSong and RyouYoun (2005). There are many techniques other than the mixing method used to enhance dispersion of particles in polymer matrices Rong et al. (2006). Surface modification of the fillers represents one of these techniques. It can be carried out physically and chemically depending on surfactants Farzaneh (2011). Ramirez et al. (2008) studied the effect of surfactants on the electrical and mechanical properties of silicone rubber. The results showed that surfactants largely affected dispersion of particles. They also observed that concentration of surfactants influenced on the matrix adsorption. As such, adsorption properties of matrix decreased with increasing concentration of surfactant in the matrix. They concluded that balance between matrix adsorption and filler dispersion was much important.

Depending on a literature survey, an enhancement in the properties of silicone rubber such as thermal and electrical conductivity, relative permittivity and hydrophobicity property attracts industries to use SIR for high voltage insulators. An improvement in the properties of silicone rubber dielectrics can be discussed using fillers as following:

Hydrophobicity of Surface

The adhesion of solid contaminations on the surface of silicone rubber decreases with increasing hydrophobicity and anti-adhesion properties and with decreasing surface energy. Silicone rubber surface with high hydrophobicity and anti-adhesion properties and low surface energy is named self-cleaning surface. Self-cleaning surface can be used effectively to suppress contamination particles like pollution flashovers. There are many methods can be used to improve the hydrophobicity of silicone rubber. Plasma treatment, which represents one of these methods, considers an effective method to make the surface of silicone rubber a super-hydrophobic surface. This is attributed to plasma treatment decreases surface adhesiveness, surface energy and coefficient of friction Farzaneh (2011). Gao et al. (2008) utilized plasma to treat the surface of SIR Silicone rubber surface was treated using CF_4 radio frequency plasma that was used to introduce fluorine groups on the surface of SIR. They found that CF_4 plasma treatment enhanced the hydrophobicity of SIR depending on increasing the contact angle of SIR with water to reach 150° .

Electrical Conductivity

It is well known that fewer polymers are electrically conductive. As such, polymers that are used for applications such as dissipating electrostatic discharges must be conductive. It can change an electrically insulator polymer ($>10^{10}$ Wcm) to become an electrically conductive polymer ($<10^5$ Wcm) by adding electrically conductive fillers into the polymeric matrix. Polymers transform from insulators to conductors when amount of conductive fillers are enough (percolation threshold) to form a continuous network of conductive particles inside polymer. Electrical conductivity of polymeric matrix increase with increasing amount of conductive fillers LilianeBokobza (2009), Yi & Choi (1999).

Thermal Conductivity

Polymers that used for insulators suffer degradation due to heat generated by dry-band arcing that can reach to higher than $500^\circ C$ Farzaneh (2011), Meyer et al. (2004). This temperature is enough to break the backbone of silicone rubber into short polymeric chains Meyer et al. (2004). Therefore, the best solution for dry-band arcing problem by using thermally conductive polymers or adding thermally conductive fillers into polymers.

SILICONE RUBBER/MONTMORILLONITE NANOCOMPOSITES

Montmorillonite (MMT) is a hydrated alumina silicate clay. Researchers have had strong attention to develop cheap organic fillers such as organic montmorillonite (OMMT) to be used instead of aero silica that is utilised to reinforce silicone rubber (SIR) Farzaneh (2011). OMMT clays, which are more compatible with organic polymers, are utilised to enhance the mechanical properties of SIR and reduce the cost Wang et al. (1998), Yang et al. (2006), Wang et al. (2006), Wang and Chen (2008), Wang et al. (2009). Global markets have shown more attention to use products of SIR/OMMT nanocomposites due to their unexpected mechanical properties Farzaneh (2011).

The most common fabrication methods to produce polymer nanocomposites are in-situ polymerization (iSP), solution processing (SP) and melt compounding (MC) Umar (2014). Solution processing (SP) (solution blending or mixing) represents the best choice to fabricate the silicone rubber nanocomposites. Therefore, many researchers Simon et al. (2008), Gharavi et al. (2010), Mishra et al. (2012), Jia et al. (2012), Ismail et al. (2013), Mishra et al. (2013), Yuan et al. (2014), Zhang et al. (2016) have used solution blending to produce the SIR/OMMT nanocomposites. Depending on the literature review, silicone rubber can be dissolved in solvent (depending on the polymer matrix and filler) under controlled mechanical stirring (Figure(4)). Following this, OMMT nanoparticles can be dispersed in the same solvent under controlled (magnetic or mechanical) stirring and then sonication in order to exfoliate the layers of the nanoclay into separated layers that are easily distributed in the polymeric matrix. Solvent is evaporated at room temperature before adding the curing agent. Two roll mill or mechanical stirrer is used to mix the OMMT, silicone rubber and curing agent. After that, the mixture is poured into molds to be left without pressure at room temperature or pressed using compression molding machine with heating to get cure samples Simon et al. (2008), Gharavi et al. (2010), Mishra et al. (2012), Jia et al. (2012), Ismail et al. (2013), Mishra et al. (2013), Yuan et al. (2014), Zhang et al. (2016). The final properties of polymer nanocomposites are strongly affected by the form of nanoparticles (intercalated or exfoliated) inside polymer matrix (see Figure(5)). In the intercalated structure, polymer chains are inserted among the layers of nanoclay leading to increase the space between layers without separation to layers. In the case of an exfoliated structure, the layers of the nanoclays are separated into individual layers that are distributed throughout the polymer matrix Mishra et al. (2012).

Mechanical Properties of SIR/MMT Nanocomposites

Type and content of nanoparticles strongly affect the mechanical properties of polymer nanocomposites. As such, the mechanical properties of the SIR/OMMT nanocomposites increase with increasing OMMT content. The influence of OMMT content on the compression set and tear properties of the HTV-SIR/OMMT nanocomposites was studied by Simon et al. (2008). They used 1, 2, 5 and 10%wt. of the OMMT to reinforce the HTV-SIR. The HTV-SIR/OMMT nanocomposites showed 24% and 32% enhancement in the tear strength and compression set respectively, as compared to HTV-SIR. The HTV-SIR/OMMT nanocomposites with 2%wt. of OMMT showed the greatest improvement in the tear strength. They proposed that tear strength and compression elasticity of the HTV-SIR matrix enhanced due to adding a small amount of OMMT into the polymer matrix. Platelets of OMMT distribute the compression forces which are applied on the RTV-SIR matrix equally, consequently, the HTV-SIR matrix is able to rebound the compression forces with minimum permanent distortion. Fuad et al. (2013) investigated the effects of OMMT size on the compression set and tear properties of the RTV-SIR/OMMT nanocomposites. The size of OMMT used was 13 and 37 μ m. RTV-SIR/OMMT nanocomposites with smaller OMMT size displayed the highest compression strength. They proposed that nanocomposites with smaller OMMT size had a great interaction between OMMT and RTV-SIR matrix (see Figure.(6-a)). This can be attributed to PDMS polymer chains can easily intercalate between smaller OMMT nanoparticles resulting from the dimensions of nanoparticles is longer than polymer chain leading to enhance the compressive strength. In contract, RTV-SIR/OMMT nanocomposites with larger OMMT size showed the greatest tear strength (see Figure.(6-b)) due to OMMT with

larger size, that it makes RTV-SIR/OMMT network provides a better restriction for tear crack propagation. The influence of OMMT content on the tensile properties of the HTV-SIR/OMMT nanocomposites was studied by M. S. Hosseini et al. (2011). The concentrations of OMMT, which were used in this study, were 1, 2 and 3%wt.. They reported that as the OMMT content increased, the tensile strength and modulus of elasticity increased, and the elongation at break decreased. As such, the enhancement in the tensile strength and modulus of elasticity of the HTV-SIR/OMMT nanocomposites were 23% and 18%, respectively. In contrast, 19% reduction in the elongation at break of the HTV-SIR/OMMT nanocomposites was observed. They suggested that adding OMMT into HTV-SIR matrix reduced chemical crosslinking, and improved interfacial adhesion between nanoclay and polymer matrix resulting from uniform distribution of nanoclay into the structure of the HTV-SIR/OMMT nanocomposites. Another study conducted by Ismail et al. (2013) reached the same conclusions. They used different types (Cloisite Na⁺, Cloisite 30B and Cloisite 20A) and contents (4, 6, 8, 10, 12 phr) of OMMT. They found that the tensile strength and modulus of elasticity of the RTV-SIR/OMMT-Cloisite Na⁺ nanocomposites were greater than RTV-SIR/OMMT-Cloisite 30B nanocomposites and RTV-SIR/OMMT-Cloisite 20A nanocomposites with rising the nanoparticles content (see Figure(7)) a and b). The addition of 12 phr of Cloisite Na⁺ enhanced the tensile strength and modulus of elasticity of the RTV-SIR/OMMT-Cloisite Na⁺ nanocomposites by about 100% and 60%, respectively. They suggested that the improvement in the tensile strength and modulus of elasticity were attributed to exfoliated layers of the nanoclays in the SIR matrix. The tendency of the Cloisite Na⁺ to form a mixture of exfoliated and partially exfoliated layers was greater than other nanoclays leading to increase the stress transfer from polymeric matrix to a single layer of the nanoclays. Cloisite 30B displayed better dispersion in the RTV-SIR matrix, however, RTV-SIR/OMMT-Cloisite 30B nanocomposites did not show great tensile strength and modulus of elasticity. This was due to Cloisite 30B reduced crosslink density of the RTV-SIR/OMMT-Cloisite 30B nanocomposites resulting from organic surfactant on the surface of the Cloisite 30B creating interference that led to decrease the crosslink density. On the other hand, RTV-SIR/OMMT-Cloisite 20A nanocomposites presented a small enhancement in the tensile strength and modulus of elasticity with increasing the nanoparticles content. The tendency of the Cloisite 20A to produce an intercalated layers was higher than other nanoclays leading to reduction in the stress transfer from polymeric matrix to layers of the nanoclays as compared to exfoliated microstructure of the RTV-SIR/OMMT-Cloisite Na⁺ nanocomposites. This might be attributed to stresses transferred from polymeric matrix to nanoclay layers passing through the stacking layers resulting in reduction in the stress transfer ability from the matrix to the nanoparticles. In addition, the ability of the intercalated Cloisite 20A to reinforce polymeric matrix was low because it had smaller surface area than other nanoclays.

Thermal Properties of SIR/MMT Nanocomposites

Yang et al. (2006) studied the effect of OMMT nanoparticles on the thermal and flame retardant properties of SIR/OMMT nanocomposites. It was utilised magnesium hydroxide (MH) and red phosphorus (RP) as synergistic flame retardant additives. It was reported that SIR/OMMT Nanocomposites displayed higher thermal stabilities and flame retardant properties than SIR matrix. As such, nanocomposites with 1% wt. of OMMT showed 129°C greater the decomposition temperature than SIR matrix. This result was in agreement with previous researches Kaneko and Yoshida (2008), Mishra et al. (2012). Another study also

demonstrated that SIR/OMMT nanocomposites showed higher decomposition temperature than SIR matrix Wang et al. (1998). As such, decomposition temperature of the SIR matrix and SIR/OMMT nanocomposites were 381°C and 440°C, respectively (see Figure(8)). They suggested that there was good interaction between SIR matrix and OMMT that improved the physical and chemical crosslinks leading to promoting the decomposition temperature Wang et al. (1998). The same result was seen in another previous work.

Dielectric Properties of SIR/MMT Nanocomposites

Silicone rubber has good dielectric properties, and OMMT is used to improve these properties. The type and content of the nanofiller influence the dielectric properties of polymer nanocomposites. As OMMT content increased the dielectric properties of the SIR/OMMT nanocomposites improved Jia et al. (2012). Razzaghi-Kashani et al. (2008) investigated the effect of OMMT content on the dielectric properties of the RTV-SIR/OMMT nanocomposites. They used 2 and 5 %wt. of the OMMT to reinforce the RTV-SIR. The dielectric permittivity and dielectric loss of the RTV-SIR/OMMT nanocomposites enhanced with increasing the OMMT content (see Figure(9)) Razzaghi-Kashani et al. (2008). This result was in agreement with previous work Gharavi et al. (2010).

SUMMARY

This review discusses the types of silicone rubber and fillers that were added to silicone rubber in order to enhancing their thermal, electrical and mechanical properties. OMMT represents the most common micro/nanofillers for dielectric applications. This review shows that electric and thermal insulation and mechanical properties of SIR were improved due to adding OMMT. Tensile strength and modulus of elasticity of SIR/OMMT nanocomposites increased with increasing the content of OMMT. While, as the content of OMMT increased the elongation of SIR/OMMT nanocomposites decreased. SIR/OMMT nanocomposites with smaller OMMT size presented the greatest compression strength. In contract, the highest tear strength was observed in the SIR/OMMT nanocomposites with larger OMMT size. SIR/OMMT Nanocomposites with high content of OMMT showed higher thermal stabilities, flame retardant properties, decomposition temperature, dielectric permittivity and dielectric loss than SIR matrix.

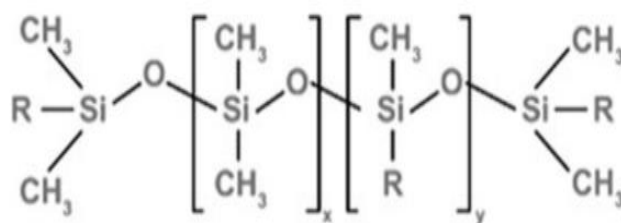


Fig. 1: Chemical structure of Polydialkylsiloxanes of silicone rubber **Shit and Shah (2013).**

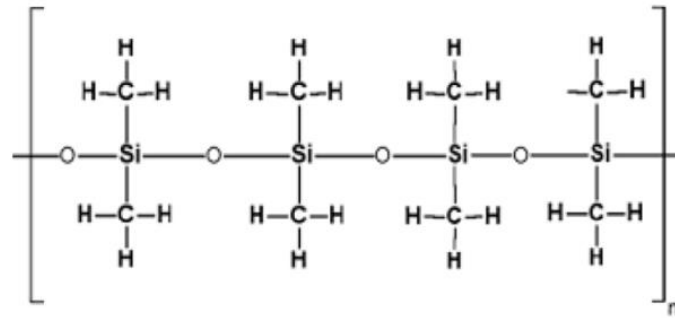


Fig. 2: Chemical structure of Polydimethylsiloxane of silicone rubber.

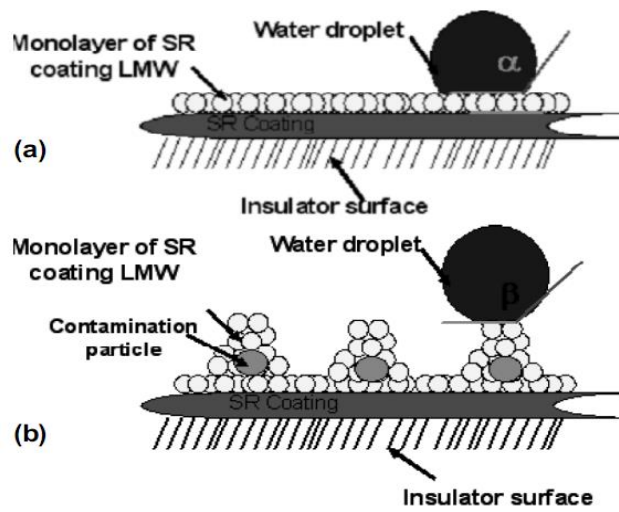


Fig. 3: Presenting silicone rubber coated insulator surface (a) without contamination particles and (b) with contamination particles Farzaneh (2011).

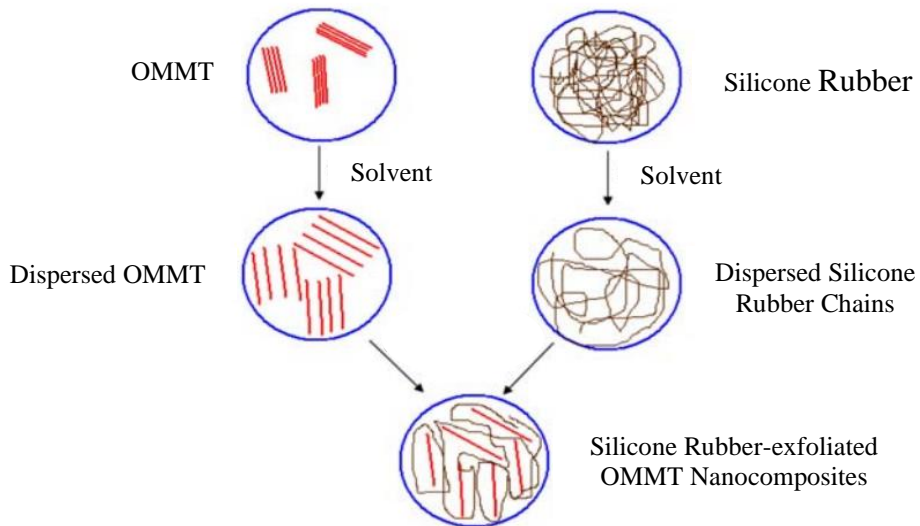


Fig. 4: Schematic of preparation of SIR/OMMT nanocomposites using solution blending method Mishra et al. (2012).

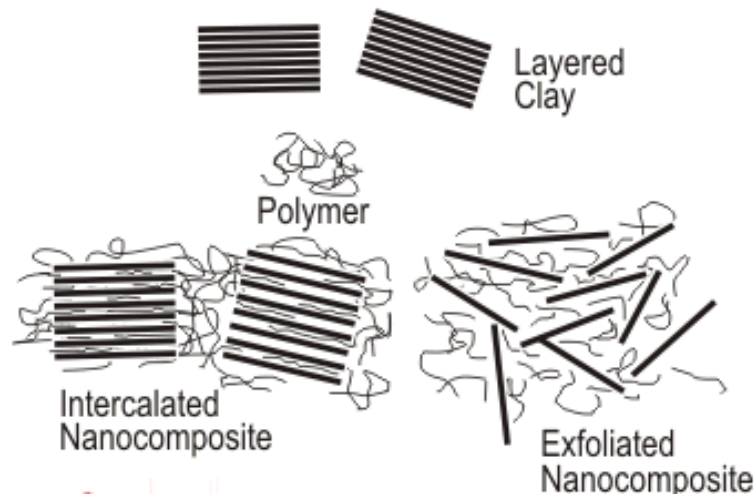


Fig. 5: Schematic of intercalated and exfoliated of polymer nanocomposites **Mishra et al. (2012)**

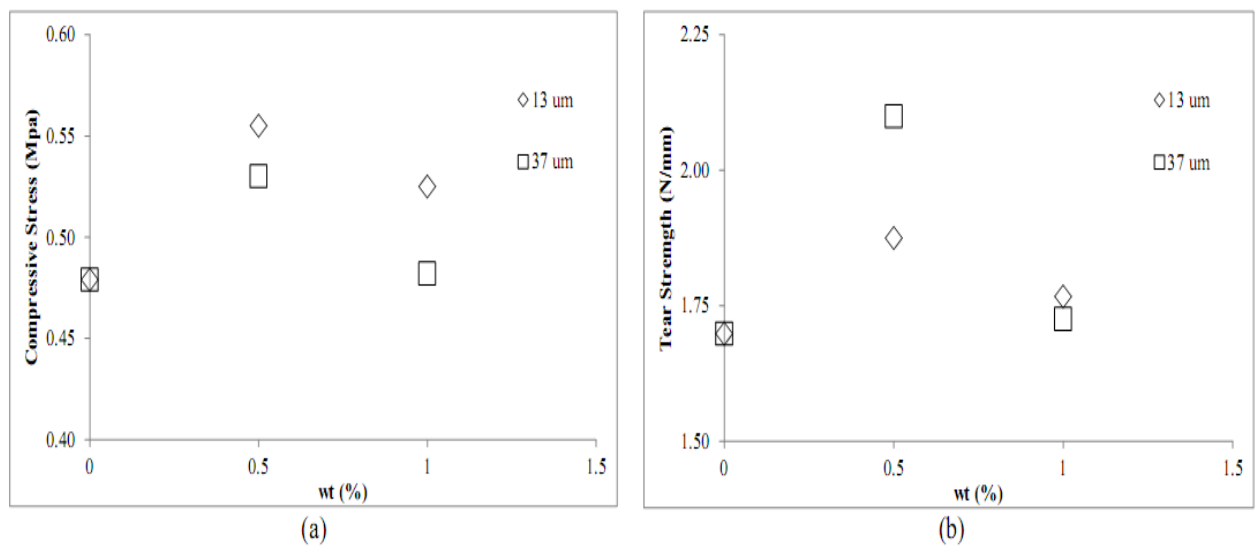


Fig. 6: Influence of OMMT size on the (a) compressive stress and (b) tear strength of the RTV-SIR/OMMT nanocomposites **Fuad et al. (2013)**

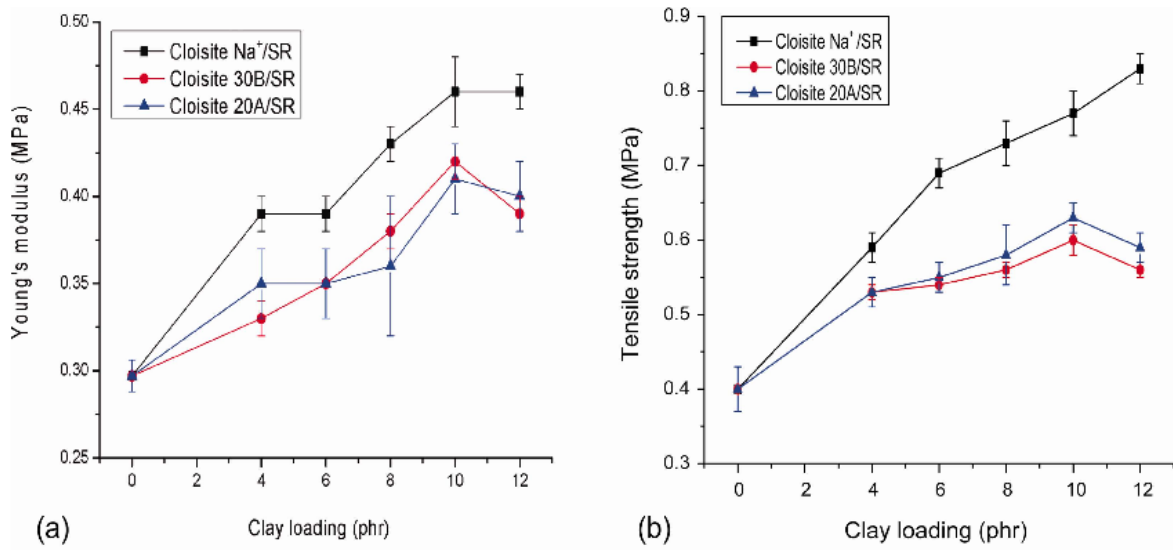


Fig. 7: Effects of type and content of nanoclay on the (a) Young's modulus and (b) tensile strength of the RTV-SIR/OMMT nanocomposites **Ismail et al. (2013)**.

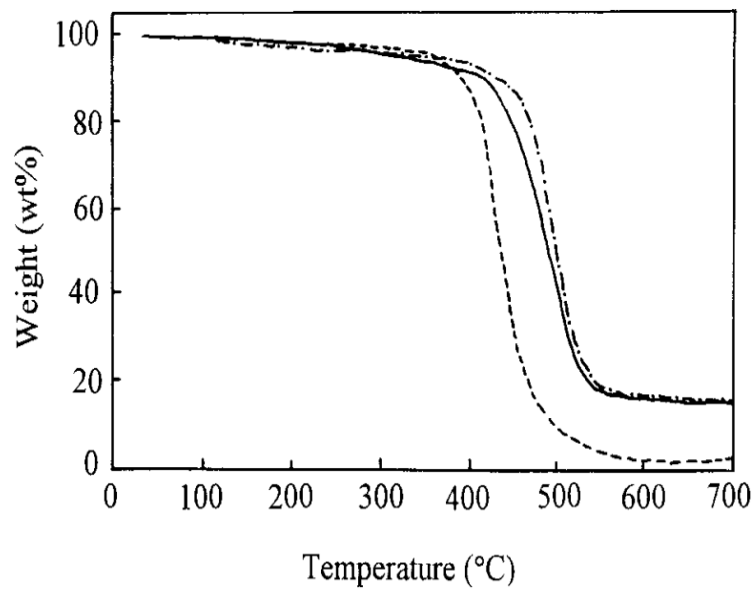


Fig. 8: TGA curves of SIR without filler (---), SIR/OMMT nanocomposites with 8.1 vol. % of filler (—), and SIR/aerosilica (— - —) **Wang et al. (1998)**.

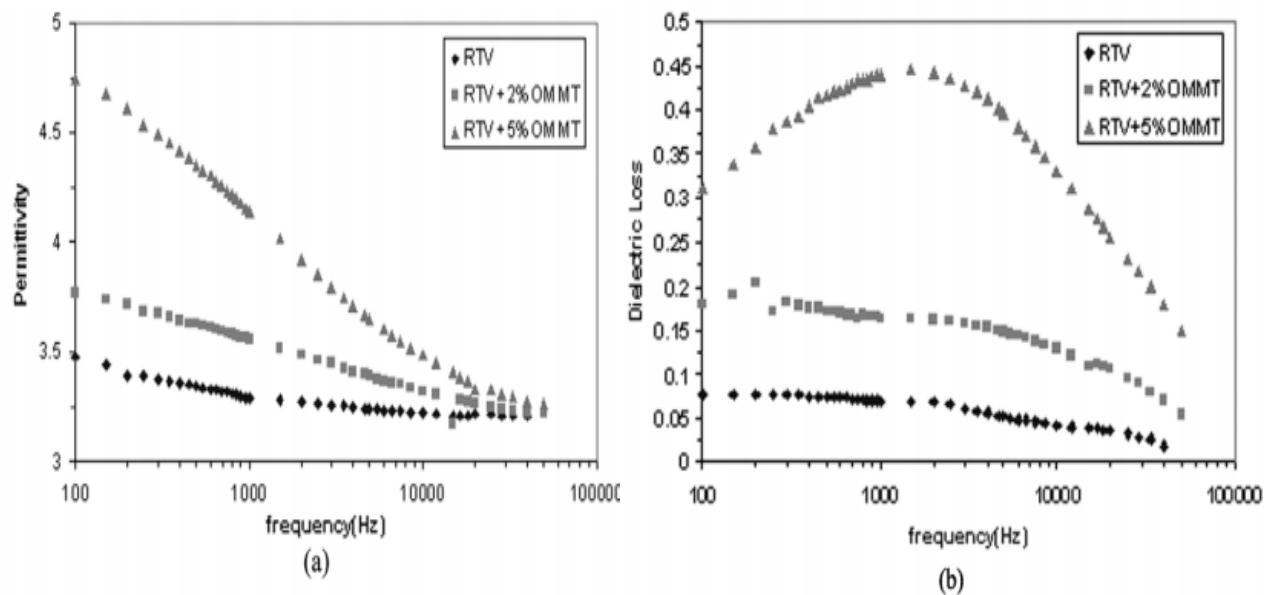


Fig. 9: Effect of OMMT content on the (a) dielectric permittivity and (b) dielectric loss of the RTV-SIR/OMMT nanocomposites Razzaghi-Kashani et al. (2008)

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