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Research paper



Mechanical and Electrical Properties of Silicone Rubber Based Composite for High Voltage Insulator Application

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Abstract

The silicone rubber (SiR) based composites has grown increasing demand in high voltage insulation application due to their exceptional ability in tackling drawbacks of existing ceramic types insulator. This paper showcase the experimental findings to understand the mechanical and electrical behavior of silicone rubber filled with various types of mineral fillers (SiO_2, $CaCO_3$, and $CaSiO_3$) specifically for high voltage insulator application. The properties variation for both mechanical and electrical attributes were analysed throughmechanical tensile testing and surface resistivity testing. Addition of mineral fillers into silicone rubber at different weight percentages (5, 10, 20, 30, 40) wt.% had caused that, as filler loading increased, the mechanical tensile performance was significantly decreased. This is due to weak reinforcing action and gap formation which found between fillers and rubber macromolecules that cause weaker cohesion between the rubber-filler components. While the value of surface resistivity is found to increase as the filler loading increased. Integration of silicone rubber with mineral fillers has resulted in no improvement of mechanical properties, but having a good surface resistivity. This interesting phenomenon was further explained by the fracture morphology evaluation via Scanning Electron Microscope (SEM) observation. From these preliminary studies, it can be concluded that, for SiR based composite system, the resulted mechanical properties and electrical properties are not interconnected between each other and both attributes are stand alone with regards to the effect of filler loading for various mineral filler types.

Keywords: *Electrical Properties;High Voltage;Mechanical Properties;Silicone Rubber; Wollastonite*

1. Introduction

Waste resources have been getting so much attention among researchers to be used as functional filler in polymer matrix in tackling few disadvantages issues. In developed countries, large amount of industrial and agricultural waste or their by-products are built up each year. The recycling of materials is of rising attention worldwide due to high environmental impact. Despite as a step towards green environment, by using waste resources also could help in improving the performance of polymer due to their special characteristics. As known, reinforcements are used as a function to improve the polymeric composite properties in terms of their electrical insulation and mechanical properties as well as to reduce the cost of resulted material [1]. Waste glass is a large component of household and industry, for example containers for food and beverages, as well as the commodity item [2]. Huge quantity of waste glass is produced daily. The recycling of glass waste materials is rising worldwide attention due to their adverse environmental impact. This has promoted the companies and researchers around the globe to develop and improve technologies to reduce or eliminating waste glass as industrial or domestic wastes, through research. This paper introduces cockle shell and waste glass as waste resources due to their extraordinary properties. Besides that, cockle shells are known for their notable mechanical and thermal properties since it is generally composed of $CaCO_3$ prismatic layers giving the shell structure a high strength, low mass and low coefficient of heat conductivity [3].

Silicone rubber (SiR) is a synthetic polymer derived from sand or quartz and has an alternating silicon-oxygen backbone, which differs from the carbon-carbon backbone in natural or organic rubbers. SiR also is known for its weatherability and ability to maintain useful properties over a wide range of temperatures [4]. It is also a hybrid that contains both inorganic and organic components. Due to this, SiR is classified as an organo-silicon compound [5]. SiR is used in the area of high voltage insulation is mainly based on polydimethylsiloxane (PDMS). PDMS is the synthesis polymer that is commonly used in SiR formulations for high voltage outdoor insulation applications, due to their characteristic of hydrophobic and water repellent [6]. The surface properties of SiR causes the recovering of hydrophobicity between contamination and corona, while other materials gradually deteriorate [7].

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Furthermore, polymer composite utilization has increased rapidly because it is one of alternative to create new materials along with good properties [8]. Other benefits employing polymer composite are their resistance to weathering, excellent surpression of leakage currents, higher flashover voltage even when wet and polluted, resistance to vandalism, higher puncture strength and the famous excellent hydrophobicity [9]. The recognition of polymer composite has grown worldwide and a large number of works have been done in order to enhance the properties of materials. Including the fabrication of new materials, the understanding of deterioration of chemical, electrical, and mechanical over the stress, design and manufacturing process of material, and also development of practical testing, monitoring, reliability methods of measuring and service performance [8]. The weight of composite insulator is about 10% of equivalent ceramic insulator. By having this advantage, it can reduce the transportation and installation costs, as well as the supporting structure that has a lower total mechanical load [10].

In the recent decades, composite insulators have been one of the major interesting research subjects due to their growing demand. Hence, there are lots of studies have been conducted to investigate the level performance of materials for high voltage insulators. Silicone rubber (SiR) has been the preferred choice for outdoor high voltage mainly due to their ability to improve electrical properties [11]. While, an advantages of composite insulator is because of their design which look a lot thinner, shorter at overall length, thus transmission lines can be more aesthetically pleasing [10]. Over the past few years, composite insulator has been found to be capable and reliable choice as near-future advanced dielectric insulation based on its outstanding properties [12]. Among their benefits, composite designs offer lighter weight, less breakage, improved seismic performance and more flexibility in design than ceramic insulators. These features can give lower installation cost, better durability and more aesthetically attractive line design. Fabricating of SiR rubber with fillers, is an increasing demand among researchers and common industrial practitioners. Basically, reinforcements material or fillers are regularly used to enhance the polymeric composite properties and also to reduce the cost of materials [8]. The common fillers that often been used for electrical insulation application are silica, calcium carbonate and wollastonite. Silicone rubber applications at high temperature for heat-resistant coatings of electrical cable were made possible. Furthermore, recent composite insulators has improved design and manufacturing techniques [13]. The following Figure 1 depicts the design and types of composites insulator that are available in commercial market. In this study, new formulation of SiR based composites will be proposed and evaluated. Effects of three different types of filler based on waste resources (SiO_2 from waste glass; $CaCO_2$ from cockle shells and $CaSiO_3$ from both glass and cockle shells) will be established along with the effects of filler loading.

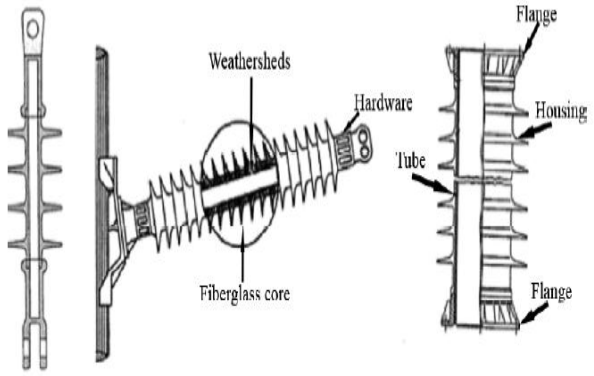


Fig. 1: Designs and types of composite insulator (14)

2. Literature Review

2.1. Mechanical Properties for SiR Based Composites

Silicone rubber (SiR) based composite has been popular among researchers due to their unique advantages. It is believed that by integrating SiR with other fillers could enhance the properties of resulted materials either in a form of mechanical or electrical properties. There have been several studies in the literature reporting on the mechanical and electrical properties, including a research done by [15]. They reported that an interaction between fillers and SiR does affecting the performance of rubber composite. Apart from that, previous research also has shown that by adding a graphene into SiR could attribute to the best improvement in mechanical properties [16]. They found an increase approximately 62% in tensile strength and 1800 s creep strain at 3.0 wt.% of graphene addition. However, the findings are contradicted with the study performed by [17] who indicated that the mechanical values are decreasing with increasing of filler percentages. It should be noted that, for different fillers which affecting the polymer matrix in a different results depending on the characteristic and integration on both filler and matrix. Apart from that, a mechanical properties also could be declined if the amount of fillers are large [12]. It has conclusively been shown that any improvements in composites are highly dependent on how good the dispersion and distribution of the fillers in the polymer matrix [18].

2.2. Electrical Properties for SiR Based Composites

On the other hand, in terms of electrical properties, the roles of fillers are believed to be used to enhance the erosion resistance of SiR, which are required for every outdoor insulation application [19]. As mentioned by [20], by adding filler to polymer matrix will enhance the performance of insulating materials. Recently, [21] revealed the effects of adding graphene nanoplatelets loadings into SiR and found an improvement on mechanical, thermal, and electrical properties. On the other hand, an improvement of tracking and erosion resistance of polymeric insulators also could be observed by adding nano-sized fillers into the base material [22]. Researchers have studied the combination of filler with Polyvinyl Chloride (PVC) would give better result when experiencing ageing process [20]. However, (18) suggesting an improvement in terms of filler dispersion as it could affect the electrical properties especially in terms of sample erosion of the sample. Previous research also confirmed that composite insulator could accelerate the decay of surface charge and increase the DC conductivity under high voltage [23]. Besides that, electrical properties of SiR/EPDM blends were found to improve most with same exposure of electron beam irradiation [24].

2.3. Fillers Used for Outdoor Insulation Application

Literature surveys shows some filler as a potential candidate for outdoor insulation application. As eloquently stated by [11], they integrated SiR with different sizes of nickel particles to be tested under various magnetic field strengths. Other than that, SiR also has been mixed with silica filler to study an erosion performance under AC voltage [25]. Besides that, calcium carbonate also has been chosen as a filler to be used to improve their performance as silicone rubber for outdoor insulation [12]. Fly ash also has been a popular material to be used as filler for insulator material for high voltage transmission tower as being utilized in a research conducted by [26]. Wollastonite is filled into Polypropylene (PP) for high voltage outdoor application [8]. However further research must be done to characterize their property in order to be feasible for commercial application. Thus, in this paper the result for high voltage insulator using different mineral fillers types integrated within SiR will be reported. The outcome is observed through tensile testing and also surface resistivity for their respective me-

chanical and electrical properties. In summary, many interesting results indicating the potential of SiR based composites have been previously reported. However, most of studies in an open literature not simultaneously examine the effect of integrating SiR with mineral fillers from waste resources. Thus, in this work, a study related to mechanical and electrical properties of SiR with various mineral fillers types at different filler loadings is presented.

3. Methodology

The main material, Silicone Rubber (SiR) was obtained from Immortal Green Industrial Sdn Bhd is used as matrix material. The type of SiR used was Elastosil R401/60S and it was bought together with an Aux Heat Stabilizer H3. Mineral fillers were added into silicone rubber which consisted of silica and calcium carbonate derived from waste glass and cockle shell respectively. Dicumyl Peroxide (DCP) acts as a curing agent to react actively with SiR with the presence of vinyl side group. Another filler name as wollastonite ($CaSiO_3$) is derived from the reaction between the silica from waste glass and calcium carbonate from the cockle shells.

3.1. Preparation of Materials

Waste glass is washed to remove any dirt and is dried in an oven operated at 80^{o}C for 24 hours to ensure all the waste glass is fully dry from water. Next, the hammer is used to break down the solid waste glass into small pieces. Then the crusher machine is utilized to make it transform into a fine size. The following process is to ensure the waste glass samples can be transformed into powder form. Then, industrial blender is used as the last step to blend the waste glass into a fine size of the powder. Cockle shell is washed to remove any dirt and is boiled using temperature 100^{o}C to eliminate bacteria. Then, it is dried in an oven operated at 80^{o}C for 24 hours to ensure all the cockle shell is fully dry from water. Next, crusher machine is used to break the cockle shell. Other than that, hammer also is used on cockle shell to make it transform into a small form. The following process is to ensure the cockle shell samples can be transformed into powder form. In this step, Horizontal Mill Machine is used with two opposing rotations. The machine is carried out at 30 min for each cycle at 400 rpm, with 15 min resting time in between. Then, industrial blender is used as the last step to blend the cockle shell into a fine size of the powder.

3.2. Preparation of Samples

The samples were prepared at different filler loading such as 5,10,20,30, and 40 % by weight. The SiR and filler along with curing agent were weighed precisely into three decimal places and mixed together using an internal mixer at room temperature with speed rotation of 70 rpm. After that, the mixture was poured into mould and compressed by using hot press machine at 175^{o}C for 20 minutes duration. The next stage was a post curing process which the materials were kept in an industrial oven at 130^{o}C for 24 hours. Few rectangular samples with dimension of 120 mm x 120 mm x 3 mm were prepared from each composition and were cut into specific size of dog bone for their mechanical tensile testing in accordance to BS 6746 and specific dimension for electrical surface resistivity testing based on ASTM D257. The following Figure 2 and Figure 3 depict the sample dimension and illustration for each tensile testing and surface resistivity testing.

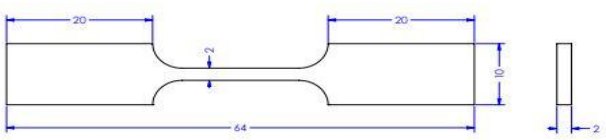


Fig.2: Sample for mechanical tensile testing

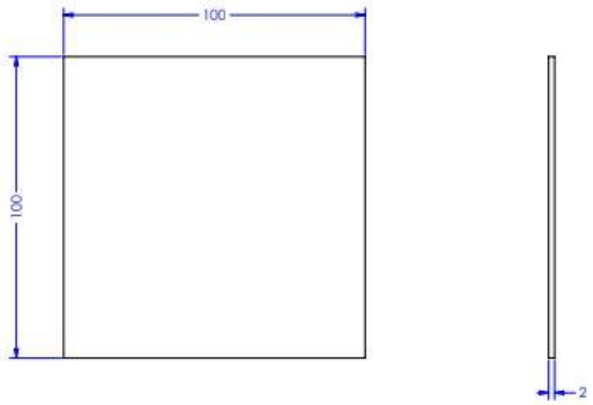


Fig.3: Sample for electrical surface resistivity testing

3.3. Mechanical Tensile Testing (BS 6746) and Surface Resistivity Testing (ASTM D257)

The tensile tests were performed using equipment that complies with ASTM Standard Test Method for silicone rubber. The standard for rubber is BS 6746 was chosen specifically for rubber materials. The tensile tests were conducted using a Universal Testing Machine (UTM) brand Shimadzu at room temperature. According to the BS 6746 standard, the specimen shall be cut into dog bone shape with dimension of 1 mm x 6.4 mm x 2mm. For this tensile machine, the gauge length used was 10 mm in length based on sample specimen. The applied crosshead speed rate of 300 mm/min is used. The average value was taken by repeating five samples for each formulation and fillers. For surface resistivity, the test was performed by following ASTM D257 and the equipment used was Monroe Portable Resistance Meter (Model 272A). The reading is taken under room temperature with humidity value ranging from 60%-70% for a period of 60 seconds. The reading collected for surface resistivity is measured in Ω/sq. The sample was cut into the dimension 10 cm x 10 cm x 2 cm (square shape) to fit into the probe size. 100 V DC was applied between main and guard electrode.

3.4. Fracture Morphological Observation via Scanning Electron Microscope (SEM)

Morphologies of composites were observed by scanning electron microscopy (SEM). All the samples were chosen by referring to the samples of mechanical tensile testing. The samples were cut into 1cm in length and sputter coated with gold prior to observation.

4. Results and Findings

4.1. Mechanical Tensile Strength Performances

As shown in Figure 4, it can be observed that, the tensile strength for all added fillers into SiR based composites are decreased as the filler loading increased. Though the tensile strength are lessened, but the values are still higher than pure SiR as control sample, which by means the tensile strength of silicone rubber based composite is as better than the unfilled SiR. The percentage of increment is about 70 % between the highest value and pure silicone rubber for the optimum tensile strength possessed by the sample 5 wt.% of SiR/$CaSiO_3$ composite. However, as the filler loading increased, the tensile strength decreased tremendously. This finding has indicated that the filler loading is not responsible for enhancing the mechanical tensile properties of produced samples.

Apart from that, this findings suggested that filler loading of 5 wt.% is even sufficient for better tensile strength.

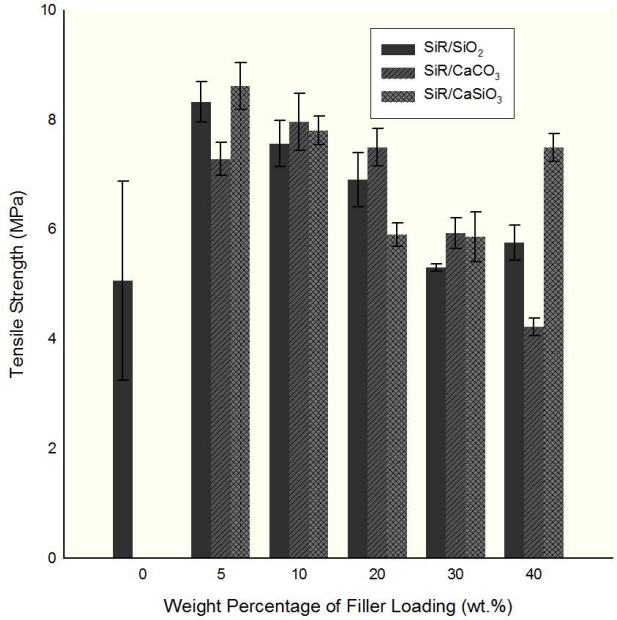


Fig. 4: Results for mechanical tensile strength behavior

This could be consistent with the findings of past studies by [22], which revealed that at lower filler concentration about 5% is enough to get better tracking/erosion resistance under DC voltages, together with other attributes. At the same time, [27] also revealed the mechanical properties could be improved even at 5 wt.% filler loading. This is due to large amount of fillers which optimumly could affects the tensile strength of insulator was limited at 5 wt.% to yield good filter-matrix interaction due to well dispersed added filler in the SiR composites. Large content of fillers could result worst agglomeration within the composite bodies which later limits the stress distribution during the tensile loading that at the end diminished the tensile performance of produced composites. The research study by [28] also found the same agglomeration phenomenon where they successfully established the correlation between filler loading and silane coupling agent towards physical characteristics of epoxidized natural rubber-alumina nanoparticles composites.

This finding also can be supported by a research done by [17] which they studied about the mechanical properties of silicone rubber-alum composites used in the manufacturing of prosthetic liners. They found that the tensile strength had decreased as the increased of alum contents due to the formation of micro gaps that acts as a defect or stress concentration area which causes the rubber not strong enough to withstand higher tensile forces. The present finding also well correlated with [29] study which concluded that if the content of inorganic filler is higher, filler particle would distort the original structure of matrix compound and resulted the physical properties of filled polymer becoming lower.

The results of present study also shows that at a filler loading of 40 wt.%, the tensile strength for both SiR/SiO_2 and $SiR/CaSiO_3$ were increased, but still lower than the value of tensile strength at 5 wt.% of filler addition. From this data, we can conclude that the fillers and matrix are dispersed well and having good bonding interaction at the highest and lowest percentage of addition only for $CaSiO_3$ filler type but not into other type of fillers. The interaction between the silica filler and polymer chains influences the mechanical strength of the polymer matrix. Silica filler with higher surface area tends to maximize the polymer/filler interaction. However, based on the above findings, silica seems not well interacted with SiR. This might due to the bonding between both fillers and matrix are not good enough causes the resulted strength of the materials decreased as the filler loading increase. Surprisingly, at 40 wt.% the strength slightly increased and was performed better than pure SiR. The surface modification might not allow the SiR chains to slip over the silica particles. $CaSiO_3$ filler sre synthetically experienced the surface diffusion between waste glass and waste cockle shell during high temperature synthesis in the furnace. This event inevitably creates surface modification or morphological changes SiO_2 and $CaCO_3$ into new phases of $CaSiO_3$ that possessed the needle-like structure. This condition might support the extraordinary performance of $SiR/CaSiO_3$ in term of their tensile strength caharacterictic. However, this result is contradicted with the finding by (30) where they found that PDMS chains able to slip over the silica particles due to surface modification. It should be noted that different materials exhibit different characteristics.

4.2. Electrical Surface Resistivity Performances

The test was conducted on all 30 samples with a portable Monroe Resistivity Meter. The results obtained were shown in Figure 5 above. The discussion of the results begin with the value of surface resistivity (SRY) obtained for all samples falls within the range of 5.3×10^{13} to 2.5×10^{14} (Ω/sq). The highest reading was spotted by sample $SiR/CaSiO_3$ at the filler loading of 40 wt% and the lowest was obtained by SiR/SiO_2 at the weight percentage of five while the rest displayed a reading ranging in between of that. Most of the readings obtained were in the range of 10^4 which signifies a high resistivity value. It should be noted that higher resistivity value indicates a better insulating materials with a small leakage [31]. This is supported by [32] study which reveal that composites filled with high filler content were highly electrically conductive. The present finding of surface resistivity obtain also consistent with findings of past studies by [33], which was about 1.37×10^{14} for the untreated SiR samples.

Based on the result, it was found that the value of surface resistivity for all the composites keep having an increment as the weight percentage of filler loading increased. It was noticed that at the filler loading 40 wt.%, there is a spike for the value of resistivity for SiR/SiO_2 fillers. This finding can conclude that only at the 40 wt.% is suitable for SiR/SiO_2 to have the best result of surface resistivity. However, the most striking result to emerge from the data is that the value of SRY for $SiR/CaSiO_3$ which shows the best value from 5 wt. % and consistent until 40 wt. %. This is due to the needle-like structure of $CaSiO_3$ which is able to enhance the electrical properties of materials. By having those results, it can be clarified that the presence of filler highly improves the value of surface resistivity of SiR samples. All the mineral fillers used in this work are successfully enhancing the electrical property of insulator by occupying an empty space within the SiR resulting in a more compact polymer with no shrinkage problems. The needle-like structure of the $CaSiO_3$ particles increase the surface area of the material caused the dispersion of $CaSiO_3$ in between the SiR particles well formed (34).

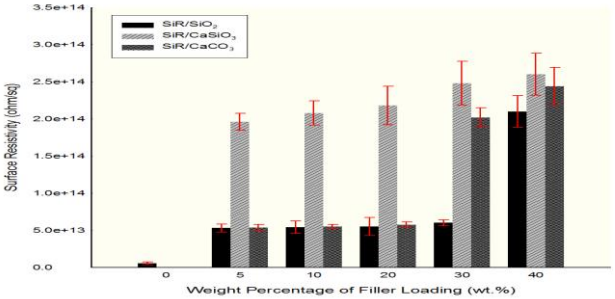


Fig. 5: Results for electrical surface resistivity testing

4.3. Scanning Electron Microscope (SEM) Observation

Scanning electron microscope (SEM) studies were performed to observe agglomeration and bonding of filler particles within the SiR matrix. Fig. 6 (a) and (b) shows the filler dispersion of $CaSiO_3$ at 5 wt.% and SiO_2 at 10 wt.% filled SiR matrix. Filler/matrix interface plays a significant role in determining both mechanical and electrical properties of the resulted SiR based composites material. In Figure 6(a), it is visually evident that the fillers are isolated and scattered uniformly over the SiR matrix, where good filler dispersion over the entire area of composites bodies are noticed. Good dispersion of fillers leads to better strength of insulative behaviour, as well as higher value of mechanical tensile properties. Sharp needle-like $CaSiO_3$ particles provide less surface energy for the filler to agglomerates between each other. This situation gives added advantage of good dispersion of $CaSiO_3$ within SiR rubber. Needle –like $CaSiO_3$ established good bonding between rubber-filler as the area of sharp tip prone to provide more surface area for matrix-filler interaction and the sharp tip also introduce the protrusion at rubber which hold the matrix during loading that permits less deformability which responsible to increase the stiffness of modulus of produced composites. However, this was not evident in composites samples that were added with silica as shown in Figure 6 (b). They were some micro crack and pores appeared in between the fracture surface of SiR matrix compared to unfilled sample at figure 6(c). The results are in good agreement with the mechanical testing results for SiR/SiO_2 composites that possessed poor tensile strength performance due to weak bonding between the filler and matrix interface. Furthermore, the interfacial bonding between constituent materials may not be strong due to void formation resulting from imperfect wetting and SiO_2 agglomeration within SiR matrix. It was clearly seen that the largest width of micro crack was 100 µm or more. The nature of circular shape of SiO_2 filler prone to cause the agglomeration phenomenon between them. This agglomeration is responsible to act as stress concentration area which prone to experience premature failure during the tensile loading which evidential from the mechanical testing results.

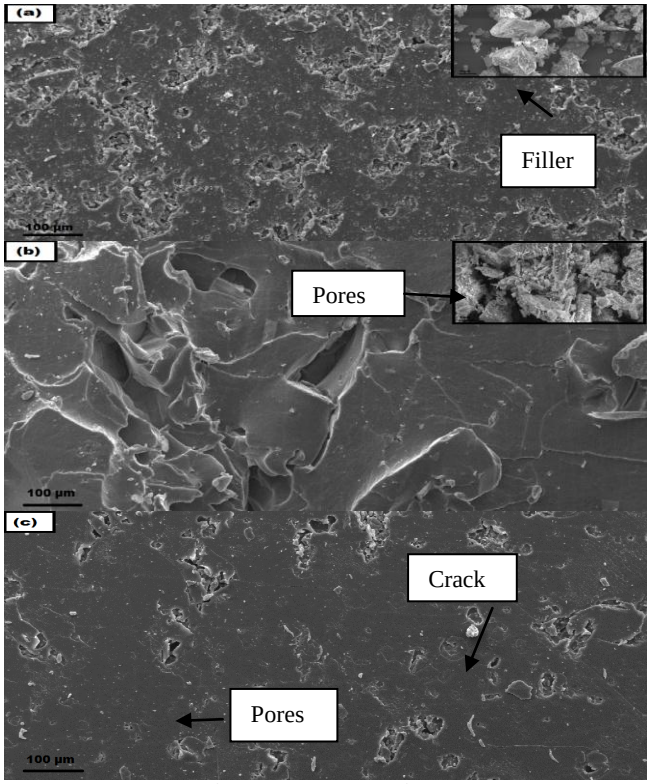


Fig.6: SEM images of silicone rubber modified by (a) wollastonite (5 wt.%) (b) silica (10 wt.%) (c) pure SiR after the tensile test (enclosed particles of wollastonite and silica at each micrographs.

5. Conclusion

As a conclusion, this study has proven that the utilization of mineral fillers from waste resources may facilitate improvements in electrical properties, but not really helping in terms of improving the mechanical properties. It should be noted that, the resulted mechanical properties and electrical properties for SiR based composite systems cannot be inter-related between each other and both attributes are stand alone with regards to the effect of filler loading and mineral filler types. For this case, the dispersion and cross-linking bonding between filler and SiR macromolecules matrix plays a significant role to achieve good performance for high voltage insulator material. Since the value of surface resistivity increasing as the filler loading increase, it can be clarified that by using the mineral fillers could make a great composite insulator. However, for mechanical tensile properties, only filler loading at 5 wt.% shows great improvement in comparison to unfilled SiR samples. This study also is successfully highlighted the role of needle-like structure of $CaSiO_3$ filler in improving both properties of interest which are the tensile strength and surface resistivity behaviour.

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