Journal of Al-Nahrain University Science Vol.15 (4), December, 2012, pp.١20-١28

Observations on the Electrical Properties of Al-Sic Composite

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Abstract

The influence of SiC particulates addition on the electrical behaviors of Al / SiC matrix composites was studied. The composited samples were prepared from Al-SiC powdered mixtures having different SiC content in the range of 40-70 wt% SiC particulates with particle sizes lower than 45- μm. The resultant powders were then uniaxially pressed in cylindrical steel die to obtain a compact disc-shaped of 1.5 cm in diameter. The compacts were sintered in the temperature range of 800 ˚C and 1100 ˚C. Electrical and dielectric properties (a.c. Conductivity, dielectric constant, dielectric loss, and dielectric loss factor) were measured for these sintered samples in the frequency range 200 Hz – 6GHz. From the results obtained it was concluded that the formation of new phases (as proved by XRD) plays a major role in the properties of these composites. Also a jump-like behavior in the measured electrical conductivity and the abrupt changes in the dielectric constant behaviors suggest frequency- dependent two- mechanisms for conduction at different SiC content.

Keywords: Al/SiC, Composite, Electric, Dielectric, X-RAY.

1. Introduction

A composite is a material made with several different constituents intimately bonded this definition is very large. A more restrictive definition is used by industries and scientists: a composite is a material that consists of constituents produced via a physical combination of pre-existing ingredient materials to obtain a new material with unique properties when compared with the monolithic material properties. This definition distinguishes a composite from other multiphase materials which are produced by bulk processes where one or more phases result from chemical composition of the metal-matrix. Al- matrix usually reinforced by Al_2O_3, SiC, C but SiO_2, B, BN, B_4C, AlN may also be considered. The aluminum matrices are in general Al-SiC, Al-Cu. As proposed by the American Aluminum Association the Aluminum Matrix Composites (AMCs) should be designated by their constituents: accepted designation of the matrix abbreviation of the reinforcement's designation arrangement and volume fraction in % with symbol of type (shape) of reinforcement [1,2].

Like all composites, aluminum-matrix composites (AMCs) are not a single material but it is a family whose stiffness, strength, density, thermal and electrical properties can be tailored. The matrix alloy, reinforcement material, volume and shape of the reinforcement, location of the reinforcement and fabrication method can all be varied to achieve required properties. The aim involved in designing metal matrix composite materials is to combine the desirable attributes of metals and ceramics. The addition of high strength, high modulus refractory particles to a ductile metal matrix produce a material whose mechanical properties are intermediate between the matrix alloy and the ceramic reinforcement. Metals have a useful combination of properties such as high strength, ductility and high temperature resistance, but sometimes have low stiffness [3].

Metal-matrix composites (MMCs) form one group of the new-engineered materials that have received considerable research since the trials by Toyota Company in the early 1980s. The most popular reinforcements are silicon carbide and alumina. Aluminum, titanium and magnesium are commonly used as the matrix phase. The density of most MMCs is approximately one third that of steel, resulting in high specific strength and stiffness. Due to these potentially attractive properties coupled with the ability to operate at high temperatures, MMCs compete with superalloys, ceramics, plastics and re-designed steel parts in several aerospace and automotive applications. The materials, however, may not have much further capacity for the inevitable future increases in service loads [4].

More recently, high volume fraction particulate aluminum matrix composite that have typical SiC loading from 40 wt% to 70 wt% have attracted considerable attention, as a result of their unique and excellent combination of properties such as high thermal conductivity, low coefficient of thermal expansion , high modulus; low density, their promising include optical mirror substrates and other optomechanical components for space based optical systems, electronic packages for avionics systems, precision components for initial guidance [5].

Silicon carbide is a promising filler material due to its good thermal and chemical stability. This is valid both during synthesis and under severe service conditions. The SiC particulates can be used with aluminum for synthesis of Al-SiC metal matrix composites. Aluminum based metal matrix composites are becoming very popular due to its lightweight, high strength to weight ratio, stiffness and wear resistance properties. The specific applications of these composites include engine blocks, pistons, brake-system components, seals, solid lubricants, wear and abrasion resistant structures, electromechanical contacts and chassis components. Owing to its high temperature electrical properties, high breakdown voltage, and high electron mobility silicon carbide is used in electronic devices that are particularly suitable for high temperature applications [6].

The research that has been carried out by A.Firdianto, et al. they investigate the influence of variation milling time aluminum (Al) with 5% silicon carbide (SiC) metal matrix composites with respect to the density, hardness, porosity, microstructure and composition of Al-SiC. The starting materials were SiC with particle size of 90 μm and Al powder with particle size 45 μm. Al-SiC powder was mixture with content of 95% aluminum (Al) and 5% SiC under process with three milling times of 1, 24 and 8 hours respectively. Powder compactness proceeded under a pressurized condition of 20 tons. The results indicate that the properties of the specimen with 24 hours milled give better properties than other milling times [7].

S. Leparoux, et al, describes the sintering of SiC-reinforced Al-matrix composites in-situ synthesis of TiC in a powder mixture of Ti and C. they noticed that the microwave energy is absorbed by SiC grains in the first case [8].

Al- SiC_p composites have been synthesized by V. C. Srivastava, et al, using spray forming process with variation in particle flow rate, size of reinforcement particles, and their volume fraction. The microstructure of composites and their electrical conductivity have been investigated. They concluded that electrical conductivity of composites decreased with increase in the volume fraction and decrease in size of the reinforcement particles [9].

An attempt were made by R N Rao et al, researchers to synthesize Al-alloy with SiC particle reinforced composite using liquid metallurgy. Microstructure, mechanical properties, and sliding wear behavior have been studded. They concluded that the composite shows a uniform distribution of SiC particles and good interface bonding between SiC particles and the metallic matrix. The sliding wear behaviors were studied using pin-on-disc apparatus against EN32 steel disc at different applied loads and constant sliding speed. The results show that the composite exhibits higher wear resistance, higher seizure pressure, less frictional heating, greater seizure temperature, and marginally lower coefficient of friction when compared with the alloy irrespective of applied load and sliding speed [10].

2. Experimental Work

2.1 Sample preparation

Al-SiC composite samples were prepared by conventional ceramic techniques. Powders of both materials with particles size lower than 45 μm were weighted using a sensitive four-digit balance type (Precisa Instruments Ltd.) then mixed thoroughly by hand in an agate mortar to produce a homogeneous mixture of composite particles. Four batches with different compositions were prepared with a different content of SiC (40 wt%, 50 wt%, 60 wt%, 70 wt %) particles. Then the weighted mass (2gm of each mixture powder) was pressed in a steel die to produce a disc-shaped sample of 1.5 cm in diameter using uniaxial pressure and 2 N for 1 min press duration. The samples were then sintered at 800°C and 1100 ± 15°C for 2 hours. The dielectric studies of composites were carried out by using a dielectric cell and an impedance analyzer (KEITHLEY Model 616). a.c. conductivity of the composites samples were evaluated.

2.2 A.C Conductivity measurements

The dielectric studies of ceramic and composites were carried out (Baghdad university, department of physics, thin film lab.) by using a dielectric cell and an impedance analyzer. (using Digital Electrometer supply type KEITHLEY Model 616). The capacitance and dielectric loss in the frequency range 200 kHz–2GHz were found out. Dielectric constant or relative permittivity were calculated by using the formula (1).

$$\varepsilon_r = \frac{c \times d}{\varepsilon_{\circ} \times A} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

Where d is the thickness of the sample, C the capacitance and A the area of cross section of the sample. ε_r is the relative permittivity of the material which is a dimensionless quantity. From these measurements, f, C, and tan δ (dielectric loss factor), for each samples were made available for the evaluation of a.c. conductivity.

Disc-shaped samples were used to find out the dielectric properties. The capacitance and dielectric loss in the frequency range 200 kHz – 6 MHz were determined. Dielectric constant or relative permittivity was calculated. The capacitance C was obtained using the analyzer and. ε_r is the relative permittivity of the material which is a dimensionless quantity.

3. Results

3-1 X- ray diffraction analysis:-

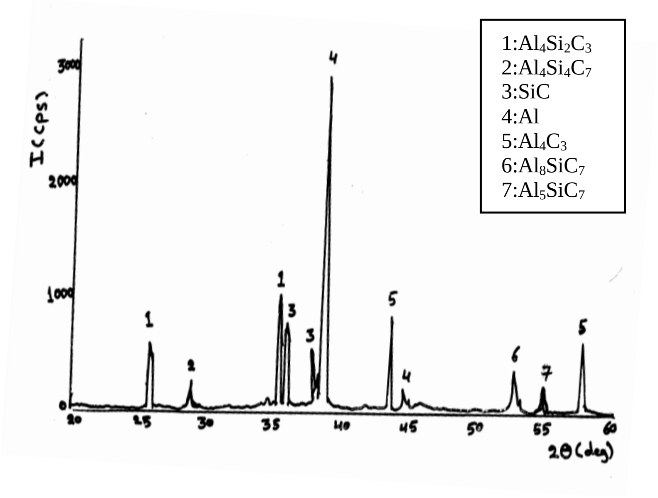

Fig.(1) Shows the x-ray diffraction pattern for sample sintered at 800 °C for 40 wt% SiC.

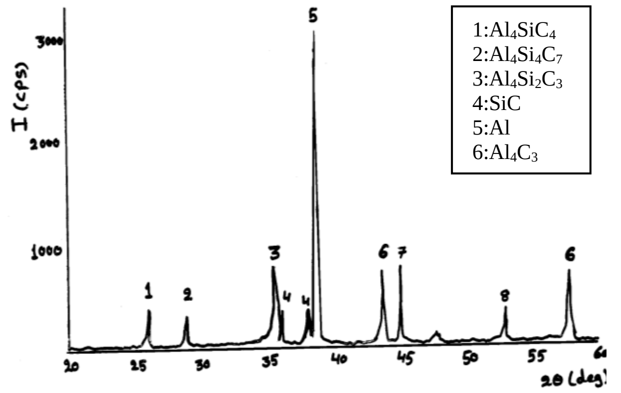

Fig. (2) Shows the x-ray diffraction pattern for sample sintered at 1100 °C for 40 wt% SiC.

To investigate the existence of any new crystalline phases in the sintered Al-SiC ceramic composites, selected samples were crushed and milled into powder using agate mortar and sieving to less than 50 micron. The powder was then analyzed by X-ray diffraction analysis (Ministry of science & technology, Iraq/Baghdad-Jadriya) using Cu-Kα radiation with a wave length 1.547 °A (Philips Ltd, Model PW 1008/85, Eindhoven, Holland). The diffracted X-ray intensity was recorded versus the diffraction angle (2θ). The 2θ angle covered the range of 10 to 60 degrees. ASTM diffraction standards were used to identify the most probable existing phases.

Fig.(1) shows the XRD analysis for Al- 40wt% SiC ceramic composite sintered at

800 ˚C for 3 hours. It was noticed that the strongest line were recorded for Al metal at the angular position 2θ=˚38.5 which have a miller indices (hkl)=(111),in addition to the presence of peaks related to SiC at the position 2θ=˚35.6 and 2θ=˚38.1 with hkl=(002) and (101). Also, analysis of the diffraction data related to Fig.(1) revealed the formation of new phases resulting from the heat treatment at this temperature. Some of these phases have a clear intensity such as Al_4SiC_4 (2θ=˚25.3), $Al_4Si^2C_5$ (2θ=˚35.3), Al4C3 (2θ=˚57.3 and 2θ=˚43.5), and Al_8SiC_7 (2θ=˚52.62). The other phases which have a lower intensity have the compositions $Al_4Si_4C_7$ (2θ=˚28.5) and Al_5SiC_7 (2θ=˚54.7). In contrast with these results, x- ray analysis for the sample sintered at 1100 ˚C for 3 hr with the same content (40wt% SiC) were also measured and shown in Fig.(2). The data revealed the generation and reduction analysis of some phases in addition to the main phases. The new phase that rising in this composite sample have a composition of $Al_4Si_3C_6$ (2θ=˚45). Phases that suffering reduction in its content were Al_4SiC_4 (2θ=˚36) and Al_4C_3 (2θ=˚43.5) while the phases that was retained the same content have a composition Al_8SiC_7 (2θ=˚52.8) and Al_4C_3 (2θ=˚57.53).

3-2 Frequency dependence

3-2-1- Dielectric properties

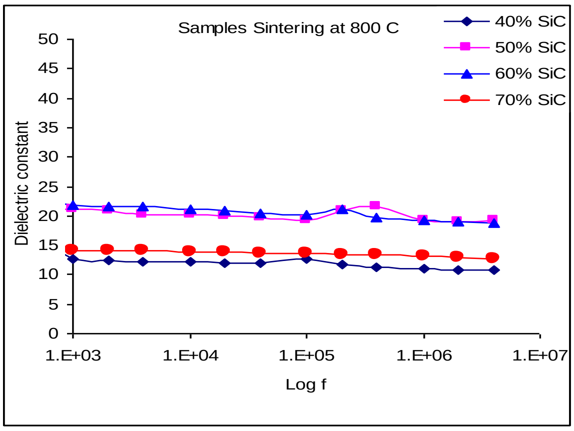

Fig.(3) The variation of dielectric constant with the applied frequency for samples sintered at 800 ˚C.

The effect of SiC addition on the dielectric constant (έ) is shown in Fig.(3). Samples sintered at 800 °C exhibited a gradually decrease of the dielectric constant with respect to frequency variation. This behavior was observed for all the prepared samples, while, the έ increased with increasing of SiC content at the same frequency range. The larger difference of the dielectric constant value in the frequency range was shown by the increase of SiC content between 40 wt% and 50 wt%, any increase of SiC content after that leads to decreasing of έ. This may be explained that the addition of SiC with specific content (50 wt% and 60 wt%) leading to the enhancement of έ, any increase or decrease of this ratio will leads to lowering of έ for the prepared samples. This may be attributed to the formation and growth of certain of SiC dielectric phases present at this specific rang content (50 wt% and 60 wt%) and exposed to specific heat treatment at 800 °C. Any change of these parameters leads to decreasing of έ.

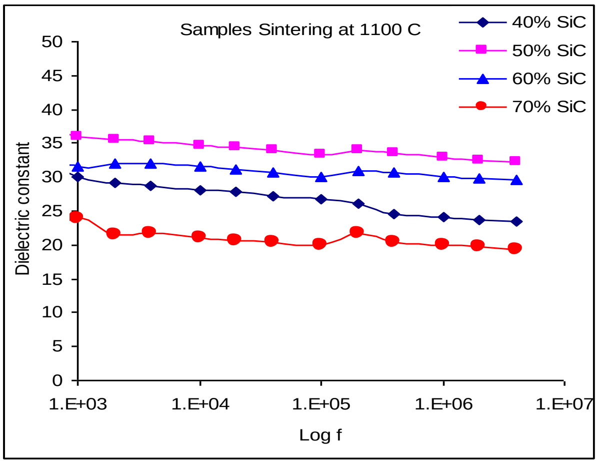

Fig.(4) The variation of dielectric constant with the applied frequency for samples sintered at 1100 ˚C.

The second group of samples which were sintered at higher temperature (1100 °C) also exhibit the same behavior over the same rang of frequency, Fig.(4). It was obviously noticed that increasing the sintering temperature leads to increasing of έ values for all the tested samples together with decreasing the differences of constant value between samples with respect to frequency variation.

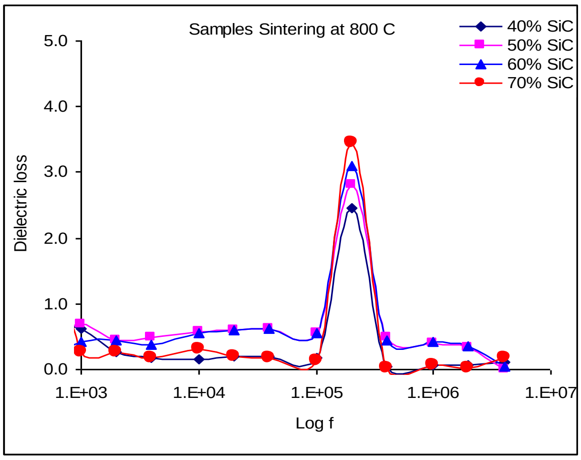

Fig.(5) The effect of SiC content on loss factor for samples sintered at 800 °C.

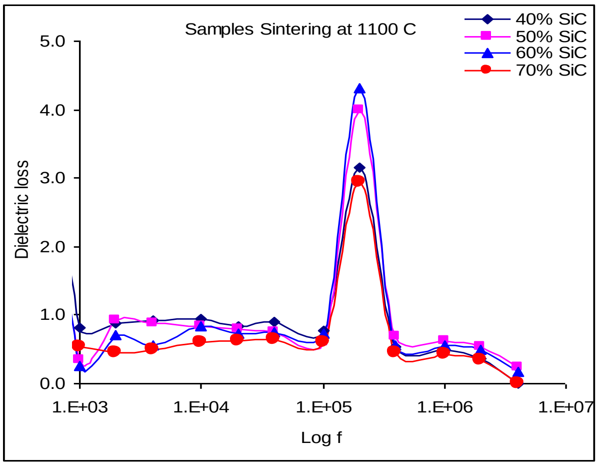

Fig.(6) The effect of SiC content on loss factor for samples sintered at 1100 °C.

The variation of dielectric loss ε″ over all the frequency rang at a different SiC content have been recorded, Figs. (5 and 6) which clearly revealed that ε″ was frequency-independent, over all the range of frequencies. Generally, the Al-SiC ceramic composite have a small value of ε″ (close to zero) with respect to variation of SiC content. Interesting phenomena have been observed in these plots. A clear loss-peaks have been occurred at a definite frequency equal to~200 kHz for all prepared samples. Samples show strong low- frequency dispersion due to space polarization accompanied by interfacial charging at grain boundaries. The ε″ is a measure of both the friction associated with changing polarization and the drift of conduction charges.

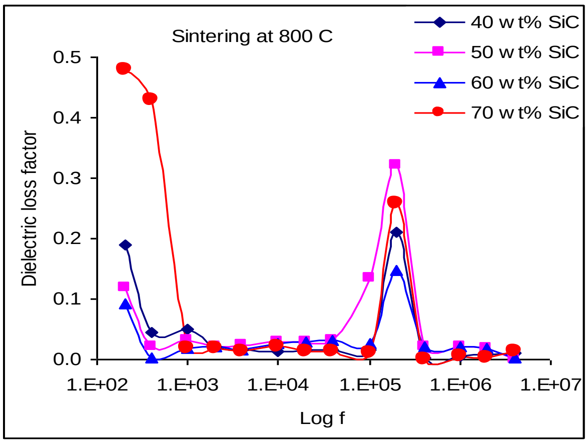

Fig.(7) The variation of dielectric loss factor with the applied frequency for samples sintered at 800 °C.

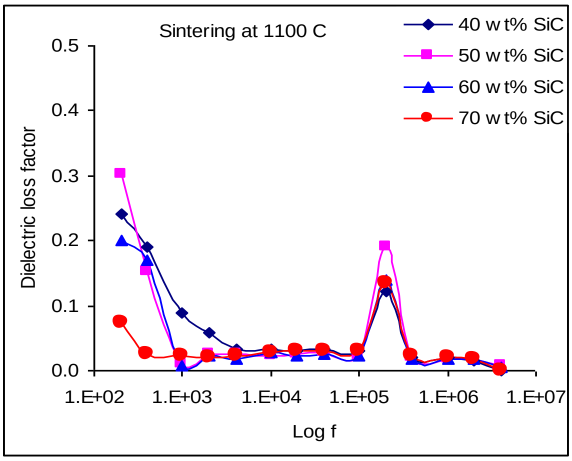

Fig.(8) The variation of dielectric loss factor with the applied frequency for samples sintered at 1100 °C.

The effects of SiC content on the dielectric loss factor (tan δ) with respect to frequency variation have been measured as shown in Fig.(7). At the low frequency, the measuring data shows that tan δ decreasing with increasing of frequency. Further increase of frequency resulted in a decrease of tanδ till it reach the same specific frequency (200 kHz) that was mentioned where the loss peak has been observed. Similar behavior was also exhibited by the samples sintered at 1100 °C as shown in Fig.(8).

4-2-2 a.c. Conductivity

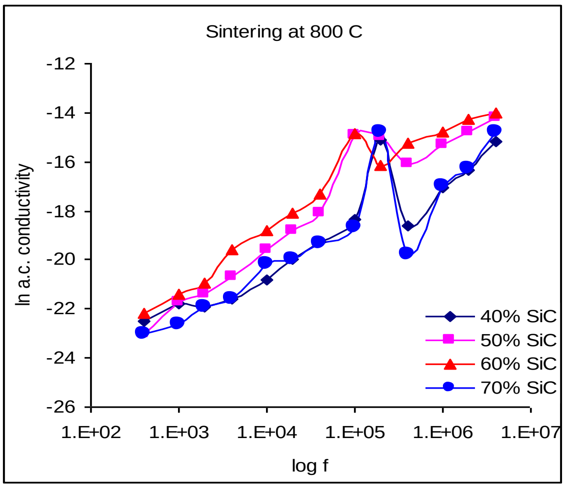

Fig.(9) Shows the dependence of a.c. electrical conductivity on frequency for representative Al-SiC composites sintered at 800 C.

The a.c. electrical conductivity of Al-SiC sintered composites has been measured over the frequencies rang (200 Hz to 6 MHz) and also at different sintering temperatures (800 ˚C and 1100 ˚C). It was observed that the a.c. conductivity increased for the prepared samples with increasing of SiC content and frequency. This is the same results obtained from previous workers (6, 12). An interesting observation has been obtained for the samples that have a definite SiC contain (40 wt% and 70 wt% SiC), In spite of the difference in SiC content, however, both have a closely measured data of a.c. conductivity. The results do not clearly understood, but it may be explained by the presence of similar phases at the sintering temperatures. These phases may inhibit the electrical conductivity. At higher frequency, samples show a distinguished peak. These peaks were located at 400 kHz for samples prepared with 40 wt% and 70 wt% of SiC. Other samples show their peaks at definite frequency equal to 200 kHz. Frequency dependence of the conductivity also shows other jump-like behaviors after 1 MHz. It was suggested that the drifting of electron was responsible for conduction in Al matrix. Also electrons and holes hopping contribute to electric conduction in SiC semiconducting particulates. The frequency dependence can be explained with the help of Maxwell–Wagner two-layer model or the heterogeneous model of the polycrystalline structure (Koops 1951). According to this theory two layers forme dielectric structure. The first layer which forms the grain boundaries consists of fairly well conducting Al-grains which surrounds layer of low conducting SiC grains. These grains are more active at low frequencies, hence the hopping frequency of electron. As the frequency of the applied field increases, the conductive grains become more active by promoting the hopping of electron, thereby increasing the hopping frequency. Thus we observe a gradual increase in conductivity with frequency. But at higher frequencies the frequency of hopping could not follow the applied field frequency and it lags behind it. This causes a dip in conductivity at higher frequencies. It was observed that a.c. conductivity for all ceramic composites increased at higher frequency (13, 14).

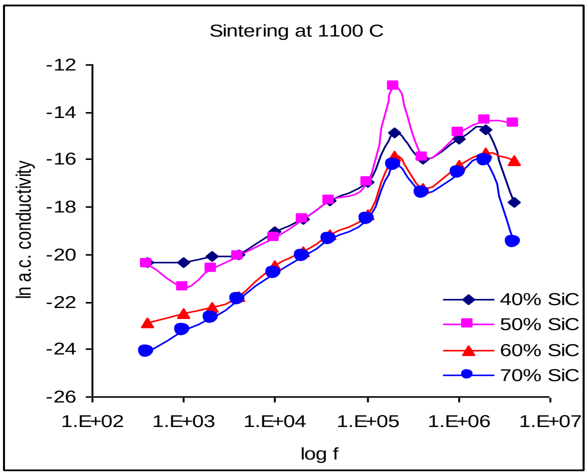

Fig.(10) Show the variation of conductivity with frequency for Al-SiC composite sintered at 1100 C.

Fig.(10) shows the variation of a.c. conductivity vs frequency for samples sintered at 1100 C. The measured data reveals an enhancing in a.c. conductivity for the tested sample that contain 40 wt% SiC, this may be due to the changes in nature of phases present in these samples as shown in the x-ray diffraction analysis. The influence of temperature on conductivity can be explained by considering the mobility of charge carriers responsible for hopping. As temperature increases the mobility of hopping electrons and holes also increases, thereby, increasing conductivity. The electrons which are involved in hopping are responsible for electronic polarization in these composites. Sample contain 60 wt% SiC shows a lower a.c. conductivity over the rang of frequency as compared with those containing 40 wt% SiC. Also the sample with 70 wt% SiC displayed a similar behavior. The limited effects of heat treatment process with increasing of SiC content may contribute to the above-mentioned behaviors. For all samples, the increases in frequency leads to increasing in a.c. conductivity until it reach its higher value at frequency 200 kHz. Further increase of frequencies reduces the conductivity. This decrease in conductivity at higher frequency is due to the inherent of carriers' motion of a.c. conductivity.

4-2-3 Compositional dependence

The variations of a.c. conductivity with Al- SiC composites at different frequencies were measured. It was found that the a.c. electrical conductivity for all prepared samples that sintered at 800 ˚C and 1100 ˚C increases with increasing frequency, as in Figs. (11, 12). Initially, increase of SiC content leads to increasing in a.c. conductivity, further increasing in SiC content revel decreasing the electrical conductivity. These results have been obtained with other workers research (9, 11). The best values have been obtained with sample content 50 wt% SiC.

This decrease in conductivity can be explained by considering the grain size of new phases formation. It was well known that the growth of grain and the formation of grain boundaries also influence the conductivity. As well as, the formation of oxygen rich layers on the surface of the grains and grain boundaries is possible. These non conducting layers increase the resistivity thereby decreasing the conductivity. The increasing of conductivity for all sintering samples with increasing frequency is due to increasing of energy for carriers current. Thus electron hopping became predominant and it increases the conductivity.

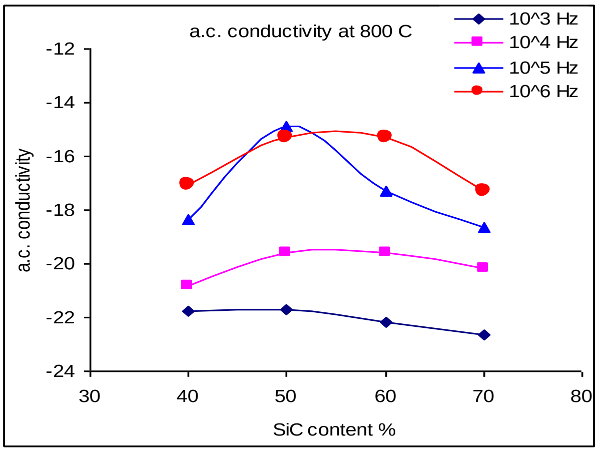

Fig.(11) Shows the variation of a.c. conductivity versus SiC wt% content at different frequencies for the samples sintered at 800 ˚C.

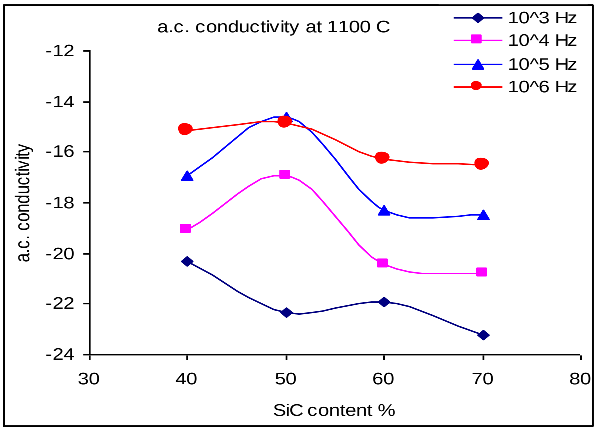

Fig.(12) Shows the variation of a.c. conductivity versus SiC wt% content at different frequencies for the samples sintered at 1100 ˚C.

5. Conclusion

1) New phases formed in the interfaces and characterized by XRD analysis are expected to play a major role in the electrical behaviors of the Al-SiC composites.
2) The most reasonable values of a.c. conductivity lies in around 50 wt% SiC in which a maxima was displayed.
3) A jump-like behaviors for the a.c. conductivity which was noticed for all the composite samples tested at different frequencies suggests frequency-dependent two-mechanisms for conduction in such composites.

4) The dielectric constant follow a semi-steady state behavior over the whole frequency range with certain abnormalities at some frequencies.
5) The higher values of the dielectric constant at higher sintering temperature may be related to the newly formed phases in addition to the possibility of matrix oxidation at the interfaces.
6) A dip in conductivity at higher frequencies may lead to the conclusion that the frequency of hopping could not follow the applied frequency.

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الخلاصة

تمت دراسة السلوك الكهربائي لتأثير اضافة دقائق كربيد السلكون على الخواص الكهربائية لمتراكبات من الالمنيوم / كربيد السلكون. حضرتَ العينات المتراكبة من خلط مسحوق الالمنيوم وكربيد السلكون بتراكيز مختلفة من دقائق كربيد السلكون وبنسب وزنية مئوية بين ٤٠ الى ٧٠ وبحجم حبيبي اقل من ٤٥ مايكرون.كبس المسحوق الناتج محوريا بوساطة قالب اسطواني من الفولاذ للحصول على اقراص مضغوطة وبقطر ١,٥ سم.لبدًَت العينات المكبوسة بمدى حراري بين ٨٠٠ و ١١٠٠ درجة

مئوية.قيست الخواص الكهربائية والعزلية (التوصيلية المتناوبة، ثابت العزل، فقدان العزل، وعامل الفقدان العزلي) لهذه العينات الملبدة في المدى الترددي بين ٢٠٠ كيلو هرتز و ٦ كيكا هرتز. من تحليل النتائج المستحصلة، تم الاستنتاج بأن نمو اطوار جديدة (كما تم اثباتها بوساطة حيود الاشعة السينية) لعب دورا" مهما" في خصائص المتراكبات. ان السلوك الشبيه بالقمة في تصرف التوصيلية الكهربائية المقاسة والتغيرات المفاجئة في سلوكيات ثابت العزل التي اعتمدت على التردد دفعت للاستنتاج بوجود ميكانيكيتين معتمدة على التردد لآلية التوصيل وعند النسب المئوية المختلفة من كربيد السلكون، متراكب، خواص العزل، خواص كهربائية، اشعة سينية Al/SiC.