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ENHANCEMENT OF MICROSTRUCTURE AND THERMAL EXPANSION COEFFICIENT OF AL/NI-SIC COMPOSITE PREPARED BY POWDER METALLURGY TECHNIQUE

Shimaa A. Abolkassem,[a] Omayma A. Elkady,[a] Ayman H. Elsayed,[a] Rabab M. Aboshahba,[b] Walaa A. Hussein[b].

[a]*Powder Technology Division, Manufacturing Technology Department, Central Metallurgical R & D Institute P.O. 87 Helwan 11421 Cairo, Egypt.*

[b] Chemistry Department, Faculty of Science, Al-Azhar University Cairo, Egypt.

ABSTRACT

Al/Ni-SiC composite was prepared via powder metallurgy technique, in which SiC particles were mixed with 10 wt % nickel powder for 24 hrs, then mixed by three percents 5, 10 and 15 wt.% with Al powder in a ball mill for 5 hrs. Three types of sintering techniques were used to prepare the composite. Uniaxial cold compacted samples were sintered in a vacuum furnace at 600^{o}C for 1 hr. The second group was the vacuum sintered samples which were post-processed by hot isostatic press (HIP) at 600^{o}C for 1hr under 190 MPa pressure. The third one was the hot pressed samples that were consolidated at 550^{o}C under the uniaxial pressure of 840 MPa. The results showed that the hot compacted samples have the highest densification values (97-100%), followed by the HIP samples (94-98%), then the vacuum sintered ones (91-97%). The XRD analysis indicated the presence of peaks correspond to Al and Ni, also Al_3Ni intermetallic and $Al_4Si_2C_5$ phases which indicated that all SiC particles were consumed with Al particles to form $Al_4Si_2C_5$. Hardness results revealed that hot compacted samples have the highest hardness values. Also the CTE results referred the decreasing of its value by increasing Ni/SiC percent.

Key words: Powder technology, Al matrix composite, Microstructure, Coefficient of thermal expansion,

1. INTRODUCTION

With increasing power density and minimization of electronic components, the need for developing efficient thermal management materials with a high thermal conductivity (TC) and a low coefficient of thermal expansion (CTE) has become urgent [1]. Particulate composites with aluminum matrix have received much recognition in applications requiring electronic packaging and thermal management. Their flexible fabrication techniques and tailorable thermo-physical properties make these composites suitable for applications such as power module base plates, microprocessor lids and printed wiring board cores. due to the largely different CTE of the ceramic particle and the metal matrix. The overall CTE of the composite strongly depends on the reinforcement type and content [2]. This opens the possibility of tailoring the composite CTE to fit the requirements of electronic systems. Particles with high thermal conductivities and strength, such as SiC, Si and diamond [3], are considered to be the best choice to reinforce the metal matrix. At present, many studies have been done for these kinds of particle reinforced metal matrix composites. Besides, MMCs allow net-shape fabrication and easy integration of functional components .To meet this requirement, aluminum matrix composites were reinforced with SiC and/or diamond particles [4]. The SiC/Al composite has made a first ingress into the thermal management market. The heat conduction and heat spreading capacity, however, cannot meet well with the increasing requirement in semiconductor industry. In addition to thermal properties, mechanical properties of such composites, e.g. bending strength, is also rigidly required for thermal management applications according to their functions of mechanically supporting protection and heat dissipation for electronic components [5]. Also the ductility of Al/SiC decreased

by increasing the volume fraction of SiC[6]. Therefore, metal modification on the SiC surface is often utilized to restrain interface reaction and enhances the interface combination [7, 8]. Metal-coated SiC powders are ordinarily produced through various processes, namely mechanical mixing, high energy ball milling, self propagating high temperature synthesis, reaction, and electroless plating. Among these methods, electroless plating is one of the most efficient in producing metal-coated SiC powders [9,10]. Recently, the production of Al based metal matrix composite has been successfully carried out using the powder metallurgy method to obtain Al/Ni-SiC composite with good microstructure and high densification [11]. This work aims at studying the effect of Ni-SiC additions on Al matrix composite prepared by different sintering techniques.

2. EXPERIMENTAL WORK

Aluminum and SiC powders with 98.9 and 99.98 % purities, 5-10 and 2-20 µm particle sizes were used, respectively. They were supplied by Loba Chemie Pvt.Ltd., India and Organik Kimya San, companies, respectively. Silicon carbide particles were mixed with 10 wt% nickel for 24 h. The nickel – SiC mix powder was mixed by 5, 10 and 15 wt.% with Al powder in a ball mill using 10:1 ball to powder ratio for 5 h [12]. The mixed powders were then consolidated using three routes. The first group of specimens was cold compacted at 840 MPa by unixial press, then sintered in a vacuum furnace at 600˚C for 1 hr by the heating cycle shown in Fig. 1. There are two holding steps in the cycle; one at 250˚C to remove all paraffin wax from the samples, and the other at 600˚C to complete the sintering process. For the second group, the vacuum sintered samples were then post-processed by hot isostatic press at 600˚C for 1 hr under a pressure of 190 MPa. A third group of samples were prepared by hot pressing at 840 MPa in a die heated up to 550˚C. These samples, are holded for 30 min at high temperatures then converted into high density blocks by hot pressing.

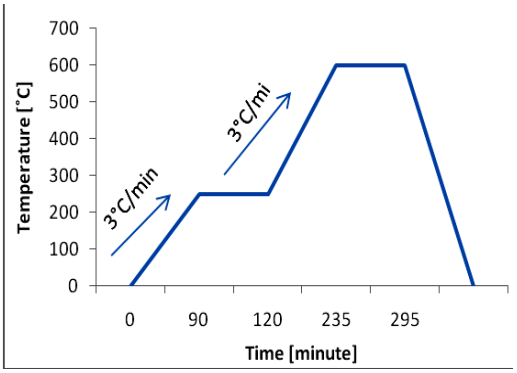

Fig . 1 Heating cycle for sintering process.

2.1. Composite characterization

The densities of sintered samples were measured according to Archimedes principle using water as a floating liquid according to MPIF standard 42, 1998, in which sintered specimens were weighed in air and in distilled water. The microstructure was investigated by scanning electron microscope (SEM; QUANTAFEG250, Holland) equipped with energy dispersive X-ray (EDX) unit, electron and backscattered electron detectors for the determination of particle size, morphology and chemical composition. The sintered samples were mounted and ground with 220,400, 800, and 1000 grid SiC papers, then polished with 3 µm alumina paste. X-ray diffraction (XRD) (model x, pert PRO PANalytical) with Cu kα radiation (λ=0.15406 nm) was used to investigate the phase structure of the sample constituents and to detect any new phases formed during the sintering process. Vickers macro hardness was measured as the average of 5 readings along polished cross section surface of the specimens by applying 3 Kg load and loading time of 15 sec. The coefficient of thermal expansion (CTE) was estimated at different temperatures from room temperature up to 350 °C for 30 min.

3. RESULTS AND DISCUSSION

3.1. Microstructure investigation

Figure. 2 shows the microstructure of Al, SiC and Ni powders. It shows that the Al particles have irregular shape like potato with particle size ranges from 5 – 10 µm. SiC has irregular shape with particle size of 2-20 µm while Ni has an irregular shape with 446 nm-1.5 µm particle size.

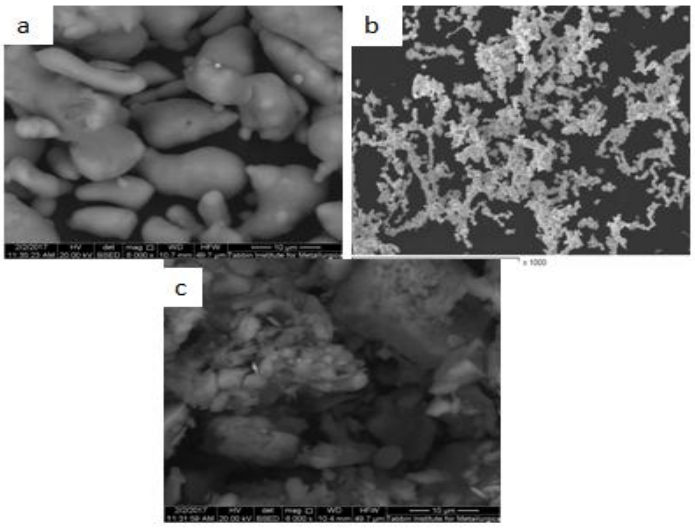

Fig. 2 SEM micrograph of (a) pure Aluminum, (b) pure SiC and (c) pure Nickel.

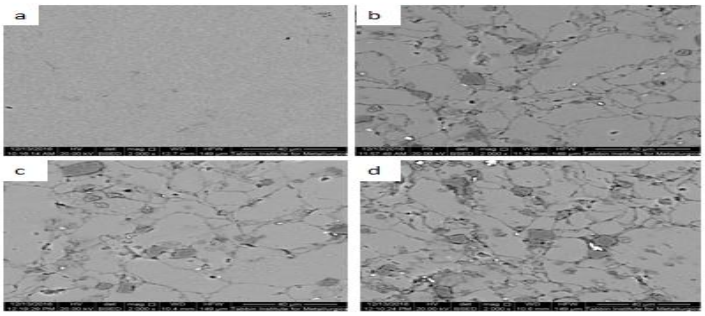

Fig. 3. SEM micrographs of (a) pure, (b) 5%, (C) 10% and (d) 15 Al/Ni-SiC for hot compacted mixed samples.

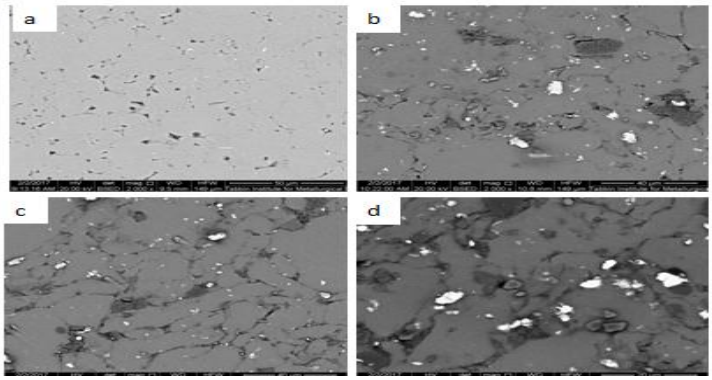

Fig. 4. SEM micrographs of (a) pure , (b) 5% , (C) 10% and (d) 15 Al/Ni-SiC for HIP mixed samples

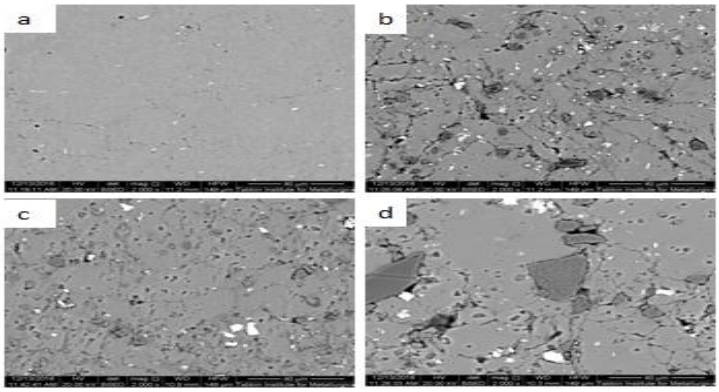

Fig. 5. SEM micrographs of (a) pure , (b) 5% , (c) 10% and (d) 15 Al/Ni-SiC for vacuum mixed samples.

Scanning Electron microscope (SEM) micrographs of pure Al and Al/Ni-SiC composite are shown in Figs. 3, 4 and 5. The EDAX analysis for the three sintering techniques are shown in Fig. 6(a,b,c). All figures show that Ni/SiC particles are homogenously distributed all over the Al-matrix. This may be due to the good mixing process and suitable mixing time of SiC with Ni which enhances the wettability between SiC and aluminum. The micrographs show four phases; one of them is the gray phase which is the Al matrix, the white spots represnt the dispersed Ni phase, and the dark gray area is SiC particles. The black spots represent the pores, that increased by increasing Ni/SiC percent. The distribution generally appeared to be fairly homogenous throughout the aluminum matrix[13]. This can be explained by the fact that,the mixed Ni/SiC particles were more easily separated from the Al matrix. By increasing Ni/ SiC content, the samples have tendency to form clusters[14] and some voids are observed due to the gaps between SiC and Al particles On the other hand, there are clear difference in the microstructure between the three consolidation techniques. In hot compacted samples all various wt% of Al/Ni-SiC nearly have the same particle size; because of fair time for grain growth. Samples in vacuum and HIP sintering, there is a good distribution for all samples containing Ni-SiC in Al matrix. There is considerable grain growth for Ni or SiC particles, are obviously visible in high wt % . It is obvious that the porosity in the composites increases with the

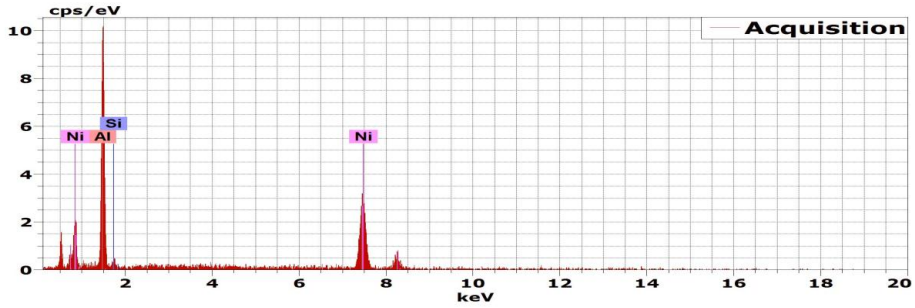

Fig. 6a. EDAX for 5% Al/Ni-SiC mixed sample by hot compacted process.

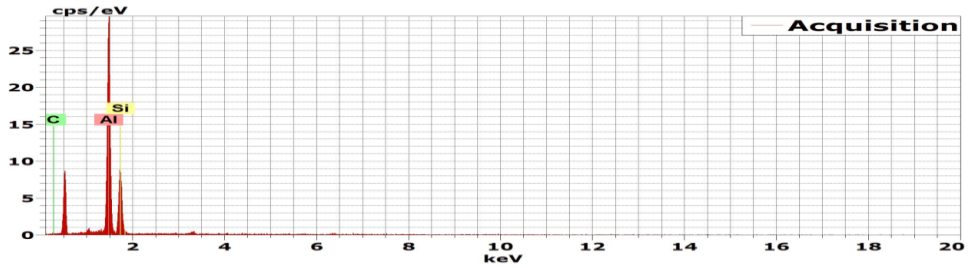

Fig.6b. EDAX for 5% Al/Ni-SiC mixed sample by HIP process.

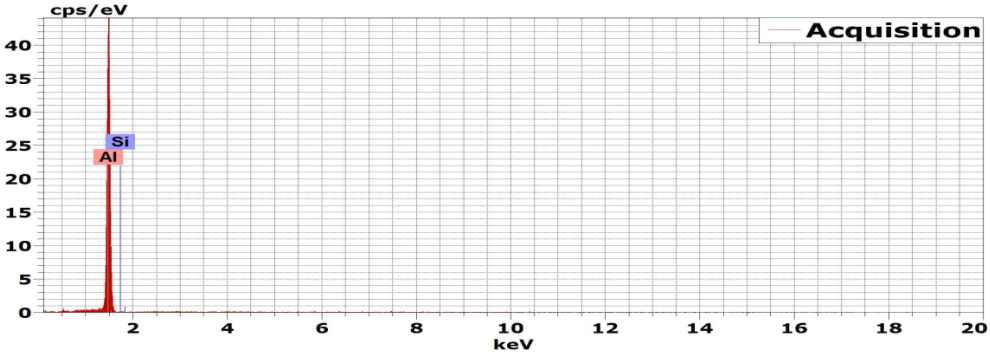

Fig. 6c. EDAX for 5% Al/Ni-SiC mixed sample by vacuum sintering process.

decrease of SiC particle sizes. The reason is that the gaps between the fine SiC particles are smaller than those between the coarse SiC particles, which makes it more difficult for Al diffusion into the matrix and fill up the gaps during the sintering process . The pores as the defect have a bad effect on the properties of the composites, such as TC, mechanical properties and relative density. It is clear that the more the pores are, the smaller the relative density is [15].

3.2. XRD study

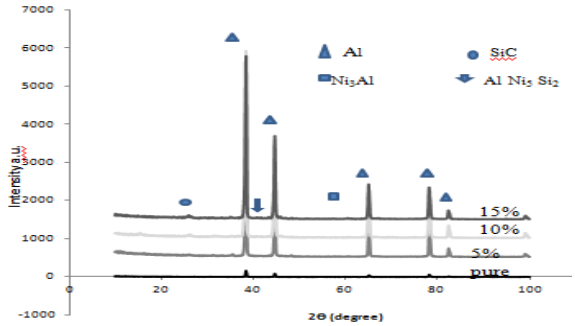

Fig. 7 XRD analysis for HIP sintering mixed samples.

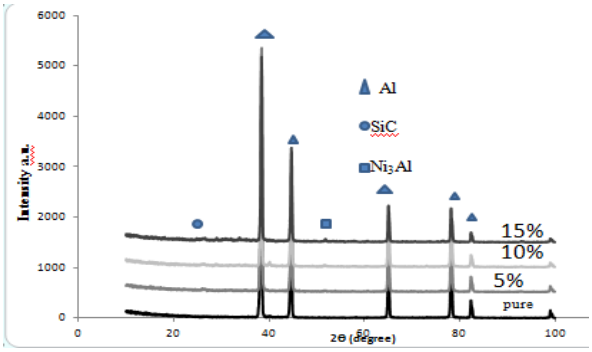

Fig. 8 XRD analysis for Hot compacted sintering mixed samples

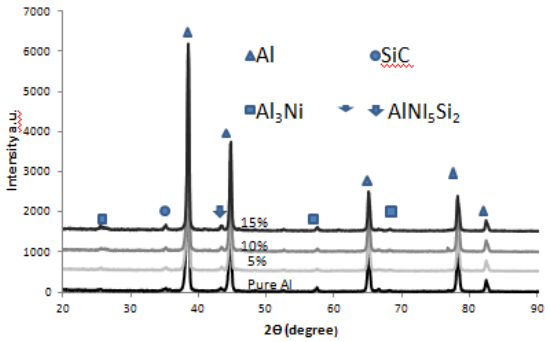

Fig. 9 XRD analysis for vacuum sintering mixed samples

The XRD patterns of Al/Ni-SiC composites which were consolidated by HIP, hot compaction and vacuum sintering techniques are shown in Figs. (7-9) respectively. It was found that there are main and sharp peaks corresponding to Al at 2Θ of 38, 45, 65, 79 and 83˚ as the major phase, traces of hexagonal SiC phase at 2Θ of 35˚ and strong peaks for Al_3Ni inter-metallic at 2Θ of 58˚ where observed, which indicates the occurrence of chemical diffusion between aluminum and nickel. The XRD diffraction of the mixed samples are shown in Figs 7 , 8 and 9. It was found the presence of four phases in case of mixed samples. Aluminum as a major phase, a clear hexagonal SiC , and both Al_3Ni and $AlNi_5Si_2$. Ni–Al intermetallic compounds (IMCs) have shown excellent properties including high melting temperature, low density, excellent corrosion resistance, and oxidation resistance. In the mixed samples strong peaks of Al and SiC were appeared because the particles of Al , SiC and Ni are free and only mixed with each other with no any bonding between them[16]. So direct reaction between Al, Ni and SiC can occur during the sintering process resulting in the formation of intermetallics such as Al_3Ni and ternary phase $AlNi_5Si_2$ according to a phase diagram reported in [17]. it can be seen that in case of hot compacted samples there were no sufficient time for the formation of $AlNi_5Si_2$ phase because the process occur in few minutes but in case of vacuum and hot isostatic synthesized samples there is sufficient time for the formation of this phase.

3.3. Density measurement

It can be seen from Fig. 11 that, there is gradually decreasing in the density value by increasing Ni/SiC percent for all samples. This can be attributed to increasing the hard SiC phase in the samples, which restricts the densification as it acts as a barrier that limits the connection between the particles leading to some agglomerations of SiC particles during the sintering process. Therefore, the capacity

for densification of the metal matrix composite decreases with increasing reinforcement content. In additions, decreasing the percentage of the deformable phase in the composite mixture results in low densification of the composite. Good densification obtained for all compositions confirm that the selected temperatures for the consolidation process were suitable for allowing the plastic flow of nickel in the Al matrix under pressure. Pure Al sintered samples are nearly fully densified. Since the compressibility of pure Al is higher than that of its composite, the densification will be higher.

Another phenomenon was observed, that the density of samples consolidated by hot compaction is higher than that consolidated by HIP furnace which in turn, is more than the vacuum samples. It is well known that at temperatures lower than 1000 °C the wettability between SiC /Al composites is poor. This may result in the presence of some pores remaining at the interface between the three phases which are adversary to samples' properties. But for the hot compacted samples, the three phases connected tightly, in the sense that nearly no pores appeared. This may be due to that fabrication of hot compaction process in which Al exists in semi liquid state when heated to some temperature and had the ability to flow freely. Meanwhile, the high pressure acts efficiently with the high temperature. The two factors

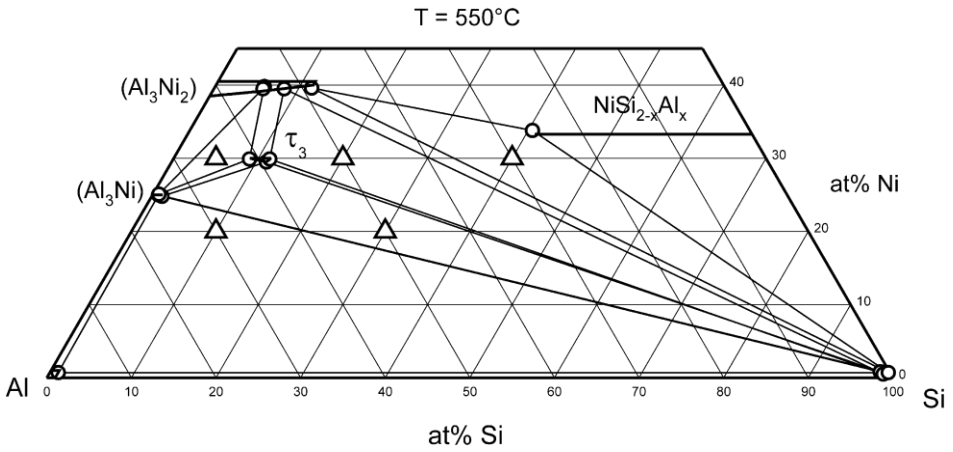

Fig. 10 Partial isothermal section of the Al–Ni–Si system at 550°C . Large triangles: nominal compositions of samples investigated by EPMA; small circles: measured phase compositions.

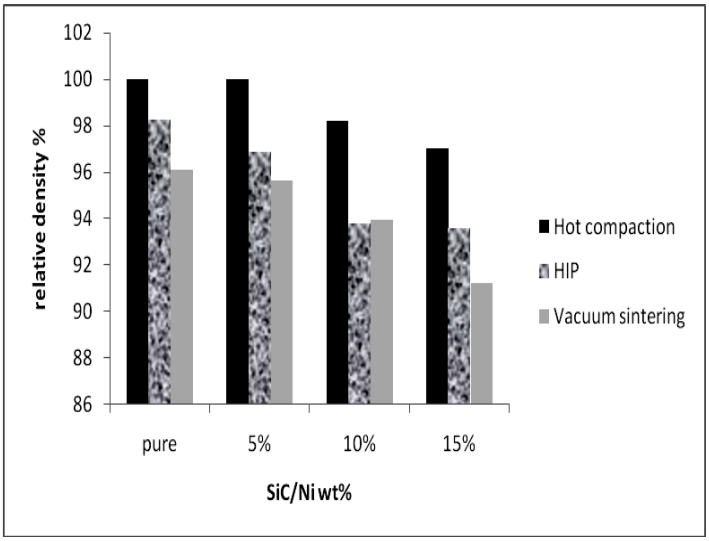

Fig. 11 Comparison between relative densities for mixed samples by three consolidation sintering techniques for pure, 5, 10 and 15 wt.% Al/ Ni-SiC.

extruded half-molten Al into the gaps of the SiC particles. Furthermore, the homogenous mixing of the raw powder in the procedure, Al in every part of the sample achieved the half liquid and half solid state nearly at the same time, and adhered SiC particles simultaneously[18].

The incomplete densification is associated with the limitation of the diffusion approach. If the particle size is smaller, the surface area increases and the diffusion is improved, and consequently the final density is closer to the theoretical one [19].

3.4. Hardness measurement

The macro hardness is a very important property that reflects the strength of the material. Generally, several factors would affect the macro hardness of the composites such as particle size, volume percent, distribution of reinforcement phase, method of preparation and density of the reinforcement [20]. The average macro hardness values of pure Al and Al/Ni-SiC composite for the three consolidation techniques are presented in Fig 12. The presence of hard SiC particles increases the macro hardness of the composites as explained by the rule of mixtures [21].

$$H_C=H_mF_m+H_rF_r \quad (1)$$

where, H_C, H_m and H_r are the hardness of the composite, matrix, and reinforcement, and F_m and F_r are the volume fraction of matrix and reinforcement, respectively. It can be seen from the figure that the hardness increases by increasing Ni-SiC wt% by nearly 15 Hv for pure sample. The increase in hardness of Al/Ni-SiC composites when compared to pure Al can also be attributed to a good interface between the soft Al phase and hard SiC phase formed during milling process and a homogenous distribution of the SiC particles in the Al matrix occurred. As the volume fraction of silicon carbide increases, the hardness value increases, by which a value of over 46 Hv was achieved for the Al-15 wt % Ni-SiC composite, [22]. This can be attributed to over helming of hard SiC particles and its constraint towards localized plastic deformation coupled with good matrix-reinforcement interfacial integrity.

3.5. Thermal Expansion Coefficient

The CTE is expressed as the change in the dimensions of the material as a function of temperature. In general, thermal expansion coefficient (CTE) of metal matrix composites (MMCs) is mainly attributed to the thermal expansion behavior of the matrix

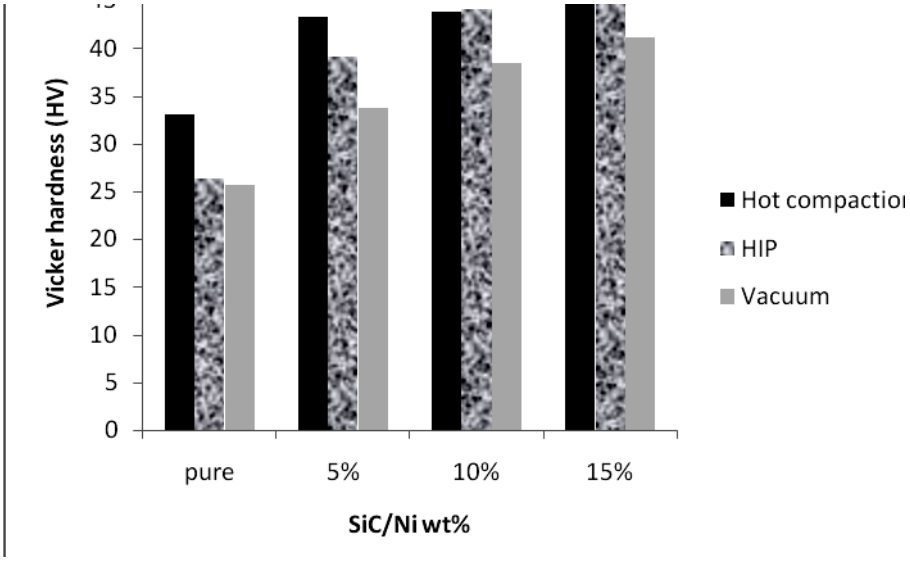

Fig. 12. Comparison between Vickers hardness for mixed samples by three consolidation sintering process for pure, 5, 10 and 15 wt.% Ni-SiC.

and reinforcement, but the pore and interfacial area have a crucial effect on the CTE of the MMCs. The thermal expansion behavior is determined by several factors, including type, morphology and volume fraction of the reinforcements, microstructure of the matrix, thermal history as well as the presence and amount of porosity. In addition, the internal stress between the matrix and the reinforcement also has a strong influence on the thermal expansion behavior[23].

It can be seen from Fig. 13. that the thermal expansion coefficient decreases by increasing SiC wt% in the three consolidation techniques, but a decline was noticed to be more than the other consolidation techniques samples observed in the samples sintered in the vacuum furnace. This phenomenon may be due to the low density of these samples compared with the others. Increase in the pores due to the lower density value that has an effect in improving thermal expansion coefficient for these samples. when the composites are heated, part of Al expands into the pores, thus limiting the increase of the bulk volume of the composites. In addition, the interface between the matrix and reinforcement can restrict the thermal expansion of the matrix. With the same SiC volume fraction, fine SiC particles have larger total surface area than coarse SiC particles, which results in a more effective restriction of the thermal expansion of the Al matrix. Therefore, the CTE of the composites of fine SiC particles is lower than that of the composites with coarse SiC particles[15].

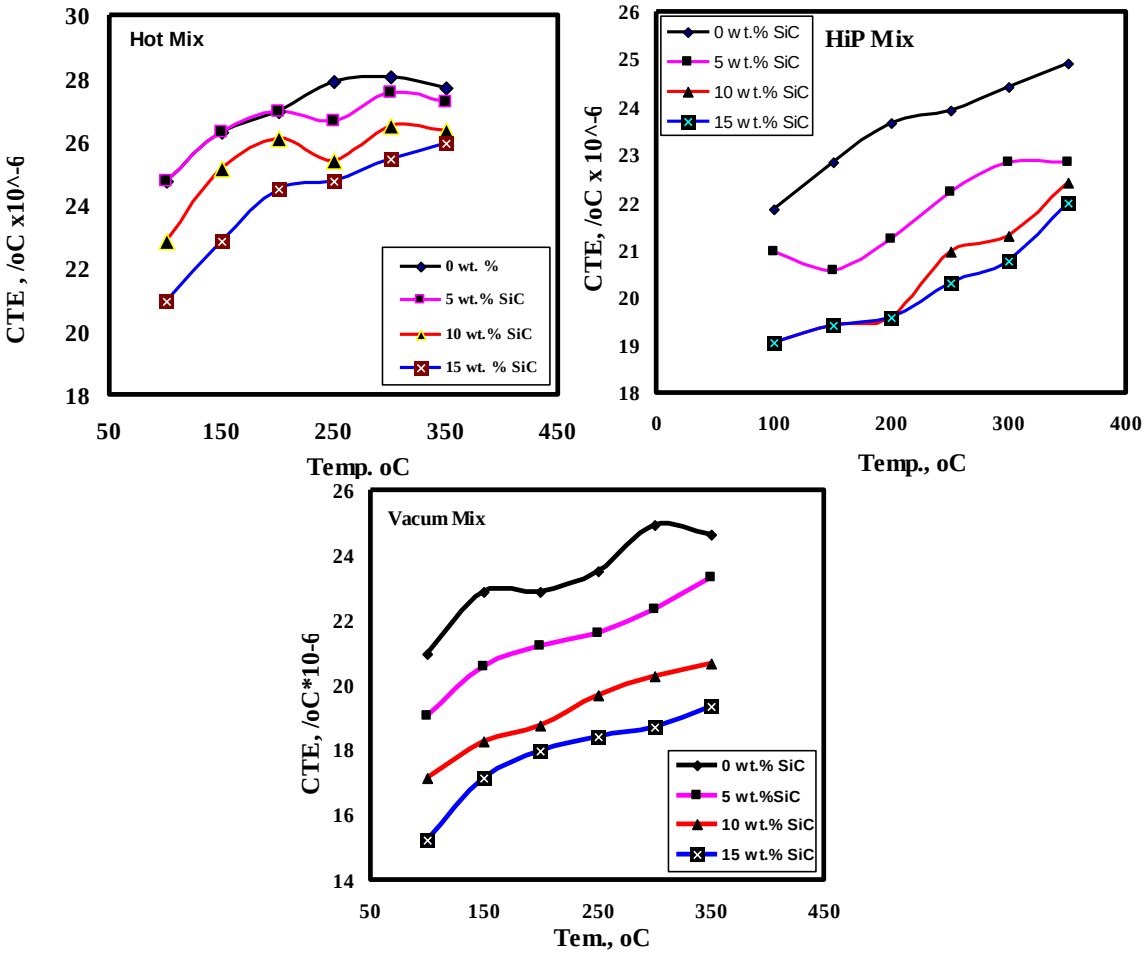

Fig. 13. Thermal expansion Coefficient for mixed samples by three consolidation sintering process for pure, 5, 10 and 15wt.% Al/ Ni-SiC.

4. CONCLUSION

1- SEM micrographs show that Ni-SiC particles are homogenously distributed over Al matrix composite.

2- All nano- Ni/SiC composites can be successfully prepared by powder metallurgy route.

3- The XRD analysis indicated the presence of Al, SiC, Ni_3Al and Ni, $AlNi_5Si_2$ for the mixed samples for the three groups.

4- The density values decrease by increasing the Ni/SiC percent for all three consolidation techniques. Samples prepared by hot compaction have the highest values than those prepared by HIP and vacuum furnace.

5- For all consolidation techniques, the hardness increased by increasing Ni/SiC percent but its value is higher in hot compaction than the other techniques.

6- Thermal expansion coefficient decreased by increasing Ni-SiC percent for all three consolidation techniques, but its value in samples sintered in vacuum furnace was more improved than other values.

REFERENCES

[1] Z. Tan, Z. Chen, G. Fan, G. Ji, J. Zhang, R. Xu, A. Shan, Z. Li, D. Zhang, Effect of particle size on the thermal and mechanical properties of aluminum composites reinforced with SiC and diamond, Materials and Design 90 (2016) 845–851.

[2] N. Sharma , R.K. Misra, S. Sharma, Modeling of thermal expansion behavior of densely packed Al/SiC composite, International Journal of Solids and Structures, 102–103 (2016) 77–88.

[3] C.X. Li, X.T. Wang, L.H. Wang, J.W. Li, H.X. Li, H.L. Zhang, Interfacial characteristic and thermal conductivity of Al/diamond composites produced by gas pressure infiltra- tion in a nitrogen atmosphere, Mater. Des. 92 (2016) 643–648.

[4] J.M. Molina-Jordá, Nano- and micro-/meso-scale engineered magnesium/diamond composites: novel materials for emerging challenges in thermal management, Acta Mater. 96 (2015) 101–110.

[5] C. Xue, H. Bai, P.F. Tao, J.W. Wang, N. Jiang, S.L. Wang, Thermal conductivity and mechanical properties of flake graphite/Al composite with a SiC nano-layer on graphite surface, Materials and Design 108 (2016) 250–258.

[6] A. Fathy, A. Sadoon, M. Abdelhameed, effect of matrix/ reinforcement particle size ratio(PSR) on the mechanical properties of extruded Al-SiC composites, Int J Adv manufacture technology, 73, (2014), 1049-1056.

[7] S.S. Zhang, K.J. Han, L. Cheng, The effect of SiC particles added in electroless Ni–P plating solution on the properties of composite coatings, Surf. Coat. Technol. 202 (2008) 2807–2812.

[8] L. Zhang, X.B. He, X.H. Qu, B.H. Duan, X. Lu, M.L. Qin, Dry sliding wear properties of high volume fraction SiC_P/Cu composites produced by pressureless infiltration, Wear 265 (2008) 1848–1856.

[9] X.M. Wu, W. Sha, Experimental study of the voids in the electroless copper deposits and the direct measurement of the void fraction based on the scanning electron mi- croscopy images, Appl. Surf. Sci. 255 (2009) 4259–4266.

[10] L.M. Luo, J. Yu, J. Luo, J. Li, Preparation and characterization of Ni-coated Cr_3C_2 powder by room temperature ultrasonic-assisted electroless plating, Ceram. Int. 36 (2010) 1989–1992

[11 A. Canakci, T. Varol, S. Ozsahin, Met. Mater. Int. 19 (2013) 519–526.

[12] B. Ryang Kim, K. DoWoo, J. Kook Yoon, Jung-Mann Doh, In-Jin Shon "Mechanical properties and rapid consolidation of binderless niobium carbide" Journal of Alloys and Compounds, V. 481 (2009) 573–576.

[13] H. M. Zakaria, microstructure and corrosion behavior of Al/SiC metal matrix composites, Ain shams engineering journal, 5(2014)831-839.

[14] F. Kertz, Z. Gacsi, J. Kovacs, T. Piezonka,the electroless deposition of nickel on SiC particles for Al matrix composites, surface and coating technology 180- 181(2004)574-579.

[15] F. Teng, K. Yu, J. Luo, H. Fang, C. Shi, Y. Dai, H. Xiong, microstructure and properties of Al-50%SiCcomposites for electronic packaging applications, Trans. Nonferrous Met. Soc. China 26(2016)2647-2652.

[16] S. Abolkasem, O. Elkady, A. Elsayed, W. Hussein, H. Yehia, effect of consolidation techniques on the properties of Al matrix composite reinforced with nano Ni-coated SiC, results in physics, 9(2018) 1102-1111.

[17] W. Klaus. richter, H. Ipser. the Al-Ni- Si phase diagram between 0 and 33.3 at .%Ni Intermetallics, 11(2003), 101-109.

[18] M. yang, D. Zhang, X. Gu, L. Zhang, effect of SiC particle size on CTEs of SiC /Al composites by pulsed electric current sintering, materials chemistry and physics 99(2006)170-173.

[19] V. Bonache, M.D. Salvador, A. Fernández, A. Borrell Int. Journal of Refractory Metals and Hard Materials 29 (2011) 202–208.

[20] S.A. Khadem, S. Nategh, H. Yoozbashizadeh, J. Alloy Compd. 509 (2011)2221–2226.

[21] R. Roy, D. Agrawal, J. Cheng, M. Mathis, Ceram. Trans. 80 (1997) 3–26.

[22] L.Y. Chen, J.Q. Xu, H. Choi, M. Pozuelo, X. Ma, S. Bhowmick, J.M. Yang, S. Mathaudhu, X.C. Li, Nature 528 (2015) 539–543.

[23] Z. Wei, P. Mai, H. Wang, C. Zou, S. Scudino, K. Song, K. Prashanth, W. Jiang, J. Eckert, the thermal expansion behavior of SiC/Al- 20Si composites solidified under high pressures, materials and design 65(2015)387-394.