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Improvement of CNFs/SiC nanocomposite properties obtained from different routes and consolidated by pulsed electric-current pressure sintering

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ABSTRACT

The influence of the preparation route and composition on carbon nanofibers-silicon carbide (CNFs/SiC) nanocomposites properties were studied. Nanopowders were mixed by ultrasonic dispersion or high attrition milling and the consolidation was done by pulsed electric-current pressure sintering technique. The relative density and fracture strength of high-energy attrition milled CNFs/SiC nanocomposites gradually increased with the increase of sintering temperature, from 1400 ºC to 1800 ºC and holding time 1 to 30 minutes. A chemical surface coating of CNFs with alumina precursor is proposed as a very effective way for improving the interaction between CNFs and SiC.

An increase of 54% in fracture strength was achieved on the nanocomposites when the surface coating was used. As a consequence of the stronger interaction between the components, which is achieved through the use of suitable processing route and sintering parameters, and the role of nano-alumina as sintering aid improved mechanical properties were achieved.

Keywords: A. Mechanical characterization, B. Ceramics, B. Nanostructured materials, D. Grain refinement, Pulsed electric-current pressure sintering

1. INTRODUCTION

Silicon carbide (SiC) is one of the most promising structural materials for high temperature applications because of its excellent strength or creep resistance at high temperature. This behavior is due to their strong covalent bond, which brings a high-decomposition temperature of 2545 ºC [1]. However, the covalent bond of SiC is responsible of its poor sinterability, showing much difficulty in fabrication of dense ceramics materials without additives. Therefore, conventional (pressureless) sintering is usually favored with sintering aids, such as the combined use of boron and carbon which were found first by Prochazka [2]. Alternatively, high-pressure sintering like hot pressing (HP) [3] and reaction sintering by immersion into melt silicon (reactive melt infiltration of silicon and its alloys into microporous carbon preforms) [4] have been utilized for the production of dense SiC.

Since the first report of carbon nanotubes (CNTs) [5] and sequentially followed carbon nanofibers (CNFs) [6], there has been increasing research and development into process of dispersing and functionalizing these materials to exploit their excellent potential as functional additives and reinforcements. Thus, carbon nanofilaments have lately

attracted remarkable attention as reinforcements of materials because of their exceptional mechanical and electrical properties [7]. About the design of composites with ceramics, research has been mainly focused on alumina-based composites [8,9], and only limited work has been done on other systems, e.g. silicon nitride or zirconia [10-12]. Despite the promising properties of this nanofilaments, there are studies showing that the addition of carbon nano-materials to ceramic matrix does not produce a reinforcement in the materials obtained [13-15]. This could be explained in terms of lack of good dispersion of CNTs (CNFs) and the poor cohesion force between the CNTs (CNFs) and the matrix [16].

In the present study pulsed electric-current pressure sintering (PECPS) or Spark Plasma Sintering (SPS) has been used for fabrication of monolithic SiC ceramic and CNFs/SiC nanocomposite materials. This sintering technique can consolidate powder compacts applying an on-off DC electric pulse under uniaxial pressure [17]. It is possible to achieve heating rates of hundreds degrees per minute, enabling the materials densification in short processing times [18].

Up to now, fabrication of homogeneous nanocomposites with carbon nanofibers and silicon carbide remains as a technical challenge. Results concerning this kind of materials have been rarely reported. Tamari et al. [19] reported the effect of spark plasma sintering on densification and mechanical properties of SiC ceramics using 280 nm SiC powder. Shimoda et al. [20] studied the preparation of CNFs/SiC nanocomposites with 1-10 wt.% of CNFs by hot-pressing via a transient eutectic phase route at 1900 ºC for 1 h, using Al_2O_3 and Y_2O_3 particles as sintering additives. They showed that the relative density of CNFs/SiC composites with CNFs content below 5 wt.% was 98% with well dispersed CNFs. However, the CNFs/SiC nanocomposites

containing 10 wt.% of CNFs possessed a relative density of 92%, accompanying CNFs agglomerates.

However, the simultaneous synthesis and consolidation of SiC using PECPS process has been only reported by Hirota et al. [21]. These authors used amorphous boron and carbon as additives and they obtained dense (~96% theoretical densities) CNFs/SiC composites with 10 vol.% of CNFs sintered at 1800 ºC for 10 min. Mechanical properties improved ~30% in comparison with monolithic SiC material.

Thus, the aim of the present contribution is to investigate the preparation of nanocomposites based on carbon nanofibers-silicon carbide by two different routes: mixture of commercial powders and sol-gel method using an alumina precursor. Sol-gel process is used to disperse alumina nanoparticles homogeneously in CNFs matrix by entrapping the dispersed CNFs in the gel network formed from Al_2O_3 precursor. The powders obtained by this route have been mixed with silicon carbide commercial powders and sintered by PECPS. The composites obtained were compared in terms of mechanical properties and microstructural features, as a function of CNFs content.

2. EXPERIMENTAL PROCEDURE

2.1. Preparation of composite powders

2.1.1. CNFs/SiC mixtures powders

The materials used in this study were commercial carbon nanofibers (CNFs) having an average outer diameter of 20-80 nm and lengths >30 μm, supplied by Group Antolin Engineering (Spain). These CNFs were generated via vapor phase growth (VGCNFs) [23] using a floating catalyst of nickel in solution (6-8%). The silicon carbide is β-SiC nano-sized powder (Hubei Minmetals Corp., China) with a mean particle diameter of 50 nm and a purity >98%. The powder mixtures were prepared by two different methods;

ultrasonic dispersion (samples labeled with US) and high-energy attrition milling (samples labeled with AM). The compositions prepared were CNFs/SiC = 100/0 - 80/20 - 50/50 - 20/80 - 0/100 vol.%. In both processing methods, the starting powders were dispersed in ethanol (Panreac Quimica). The ultrasonic dispersion was prepared in a high intensity ultrasonic horn (20 kHz, model dr. Hielscher). Time needed to achieve a good dispersion is approximately proportional to the volume of the sonicated material [24]. Therefore, the sonication time should be long enough to break up most of the nanoparticle agglomerates, but at the same time it should be kept as short as possible to avoid degradation of the material. Sonication experiments were carried out with amplitude 100%, a volume of 400 ml of the particle SiC mixture and the time employed was 10 min. High-energy attrition milling (Union Process, USA) using alumina media of 2 mm diameter was done at 400 rpm and milling times of 1 hour. The ball-to- powder ratio was 4/1. After milling, the resultant slurry was dried at 60 ºC and the dried powder was sieved under 63 μm.

2.1.2. Surface coating of CNFs

Surface coating of CNFs has been carried out employing sol-gel processing route with an alumina precursor, as it has been described in a previous work (see [34] for details). $AlCl_3 \cdot 6H_2O$ (>99% purity, Fluka) was used as alumina precursor. The modified powders have been characterized in terms of thermogravimetric behaviour and XRD analysis as shown in [34]. The final content of Al_2O_3 on the CNFs surface was 10 vol.%.

2.1.3. Mixtures powders of SiC with alumina coated CNFs

The appropriate amount of the carbon nanofibers which were surface coated with alumina precursor in the previous section were mixed with nano-sized SiC powder in order to achieve a final volume composition of 78 vol.% SiC, 20 vol.% CNFs and 2 vol.% Al_2O_3. This mixture powder was also prepared by two processing methods; ultrasonic (US) dispersion and attrition milling (AM).

2.2. PECPS and characterization of sintered bodies

The nanocomposites powder mixtures were placed into a graphite die with an inner diameter of 20 mm and cold uniaxially pressed at 30 MPa. Then, they were introduced in a pulsed electric-current pressure sintering HP D25/1 (FCT Systeme GmbH, Rauenstein, Germany) under low vacuum (10^{-1} mbar) and sintered at different temperatures; 1400, 1600 and 1800 ºC. Different dwelling times at the maximum temperature were selected from 1 to 30 min under an applied pressure of 80 MPa and a heating rate of 100 ºC min^{-1}. Bulk density of the sintered bodies was measured by the geometric method from weight and geometric volume of the material. Relative densities were calculated as relation between geometrical and theoretical densities (t.d). The fracture strength was measured using biaxial testing employing the equations of Kirstein and Woolley [25], Vitman and Pukh [26], and the standard specification ASTM F394-78 [27]. Five samples were tested for each composition. All tests were performed at room temperature using the universal machine Instron (Model 8562) with a cross-head displacement speed of 0.002 mm s^{-1}. Fracture surfaces of sintered samples were characterized by scanning electron microscopy (SEM, Zeiss DSM 950).

3. RESULTS AND DISCUSSION

3.1. Effect of sintering temperature

Mixtures of SiC/CNFs powders containing 0, 20, 40, 60, 80 and 100 vol.% of carbon nanofibers were prepared by ultrasonic dispersion. This methodology is commonly used for preparing ceramic-CNTs/CNFs mixtures [22,39]. Table 1 summarizes the relative density of the monolithic SiC and CNFs materials and CNFs/SiC nanocomposites sintered by PECPS at 1400, 1600 and 1800 ºC for 1 min of dwell time.

Although the density of monolithic SiC material increases with sintering temperature, it is lower than 75% t.d at 1800 ºC (maximum temperature tested). Concerning the influence of material composition, the density increases gradually with increasing content of carbon nanofibers and final temperature, achieving densities >90% with a 80 vol.% of CNFs. Moreover, the variation of relative density with sintering temperature is less significant for materials with high CNFs contents. It is widely known that the full densification of the SiC ceramics without sintering additives is especially complicated, mainly due to the covalent nature of the silicon carbide bond which in the sintering process shows very low diffusivity of the atoms and high energy on the grain boundaries [2]. For this reasons, sintering is normally carried out in the presence of small amounts of other substances promoting densification, referred to as sintering activators. Therefore, the use of small amounts of sintering aids, i.e. boron and carbon, have been used for sintering monolithic SiC material and CNFs/SiC composites in conventional [2], Hot-Press [28] and PECPS [21]. In order to achieve maximum densification boron and carbon are mostly introduced in the amounts of 0.5 and 3 wt.%, respectively. The idea of using these activators for silicon carbide sintering was first given by Prochazka [30] and Prochazka and Scanlan [2] yet in mid 1970s but the role of activators in the sintering process has been disputed ever since [31-33] and still remains an open question.

In this work, CNFs/SiC nanocomposites have been sintered without adding any specific sintering additive. Nevertheless, nanocomposites show significantly higher relative densities when CNFs content is increased. This trend is completely opposite to the behavior of Al_2O_3/CNFs and ZrO_2/CNFs composites [34,35], suggesting that carbon nanofibers act as sintering additive having a noticeable effect on the densification behaviour of SiC ceramics.

The fracture strength of the CNFs/SiC nanocomposites as a function of sintering temperature and volume of CNFs content are shown in Figure 1.

CNFs/SiC nanocomposites show low flexural strength values due to the high residual porosity (10-40)%, especially in the case of materials sintered at 1400 ºC. A remarkable result is that the fracture strength of nanocomposites sintered at 1600 and 1800 ºC is higher than that corresponding to their respective monolithic materials, SiC and CNFs. As it can be seen in Figure 1 the fracture strength of the monolithic SiC material is only improved when the sintering temperature is increased up to 1800 ºC while in the case of CNFs material no difference between materials sintered at 1600 ºC and 1800 ºC is observed. It has been previously shown that the optimal sintering temperature for CNFs is around 1500 ºC [36].

CNFs/SiC (20/80) vol.% nanocomposites sintered at 1600 ºC and 1800 ºC achieve a 10-20% higher fracture strength than monolithic SiC having similar or even lower relative density. This effect is more noticeable in the CNFs/SiC nanocomposite with 80 vol.% of CNFs, which shows higher fracture strength even at the lowest sintering temperature (1400 ºC). These results show a synergic effect between both phases for preparing materials with improved structural properties.

It must be noted that relative densities achieved by CNFs/SiC (80/20) vol.% and 100 vol.%, CNFs material are similar and >90% t.d when they are sintered at 1600 ºC and 1800 ºC. Nevertheless, nanocomposite sample shows higher fracture strength due to carbon matrix reinforcement by ceramic nanoparticles. The incorporation of 20 vol.% of SiC nanoparticles leads to an increase of fracture strength of carbon material from 60 to 85 MPa, that is nearly 50%, when they are sintered at 1600 ºC.

In Figure 2, they are shown the micrographs of the fracture surfaces of CNFs/SiC nanocomposites with 50 vol.% of SiC sintered at 1600 ºC and 1800 ºC. Density and fracture strength of this composite show intermediate values between CNFs/SiC (20/80 and 80/20) vol.% nanocomposites. It can be clearly observed the incomplete densification of these materials. The high level of porosity in the samples makes difficult the identification of each component. However, the irrelevant differences between both samples indicate that it is not possible to complete densification of these composites by only increasing the sintering temperature.

3.2. Effect of mixture powder method on the composites properties: ultrasonicated mixtures versus high-energy attrition milling

The homogeneous dispersion of carbon nanofibers and silicon carbide components is must if it is desired an improvement of the sintering behaviour of the composite and therefore, their mechanical properties. In order to investigate the influence of the processing method of powder mixtures on dense material properties it was explored the possibility of using high attrition milling, a technology commonly employed in ceramic processing.

The choice of the equipment for superfine and ultrafine grinding depends on the end-use of product, required product size distribution, purity requirements, etc [37]. Comparison

of the grinding performance of conventional ball mill and attrition mill has been reported and better performance of the latter was observed [38].

The attrition milled mixture powders of all the compositions studied were sintered by PECPS at 1800 ºC during 1 min of holding time. The relative density and fracture strength of the nanocomposites for these materials are shown in Figure 3, where they are compared with the results of the nanocomposites processed by ultrasonic dispersion.

No significant differences in relative density of nanocomposites prepared by both processing methods are observed. However, when nanocomposites are processed by attrition milling of powders mixtures, they show higher fracture strength. The most important differences correspond to samples with low CNFs content and for the monolithic SiC material. This result may suggest that some agglomeration problems of the silicon carbide nanopowder (50 nm) can be avoided by high energy attrition milling. Thus, CNFs/SiC (20/80) vol.% nanocomposite AM shows an improvement of 60% in fracture strength compared with the same nanocomposite obtained by US, proving the relevance of sample processing method before sintering on the composites fabrication.

In Figure 4 low-magnification SEM images of fracture surface of CNFs/SiC nanocomposites with 20 vol.% of CNFs processed by both methods are shown. Since almost 50% higher fracture strength is showed when AM process is employed instead of US to fabricate the nanocomposite, it can be concluded and observed in the right image of Figure 4 that attrition milled sample exhibits a well-dispersed carbon nanofibers in SiC matrix with homogeneous microstructure and fine grain-size. In the other hand, US sample has CNFs agglomerates being the main reason of the lower fracture strength of this nanocomposite.

3.3. Effect of holding time at maximum temperature

In previous sections it has been proved the difficulty for completing densification of SiC/CNFs composites studied in this work even at temperatures as high as 1800 ºC. Then, the following parameter studied in the sintering cycle adjustment has been the holding time. One of the most recognised features of PECPS sintering technique is the short processing times due to the high heating rates that can be applied. In order to preserve this advantage improving the material densification, a new sintering cycle was tested. Thus, the heating rate was maintained at 100 ºC min^{-1} while the holding time at maximum temperature was increased from 1 to 30 minutes. Then, the whole cycle time is around 1 hour (noticeable shorter than conventional hot-press sintering cycles). The results in terms of relative density and fracture strength of the nanocomposites and monolithic materials, processed by US mixture powders and sintered at both holding times are summarized in Figure 5.

The analysis of these results allows obtaining the following conclusions:

i) An increase in the holding time has no effect on densification of carbon nanofibers matrix materials. Thus, density of SiC/CNFs (20/80 and 0/100) vol.% materials are similar when they are sintered using 1 min or 30 min holding time. The inefficiency of increasing sintering time is due to the absence of diffusion mechanisms in carbon matrix materials. In the case of SiC/CNFs (20/80) vol.% nanocomposite, although there is a ceramic phase, its content is too low and it acts as a reinforcing component dispersed in the carbon matrix. Fracture strength results are also similar for materials prepared with both holding times.

ii) Monolithic SiC density is similar for both holding times tested. However, an important increased in its fracture strength is observed. Porosity content is not reduced due to the absence of sintering additives but a longer holding time improves the resistance of necks formed between SiC particles leading to an increase in the fracture strength.

iii) The most important improvements in density and mechanical resistance are observed for SiC/CNFs (80/20) vol.% nanocomposite. This material can be described as a ceramic (SiC) matrix reinforced with CNFs as second phase. The carbon component acts as a sintering additive and its good electrical conductivity allows a homogeneous distribution of electrical current through the whole sample. Then, carbon nanofibers are simultaneously one component of the final material and an active element enhancing the sintering process by PECPS. Although SiC/CNFs (50/50) vol.% nanocomposite also show density and fracture strength improvements, they are less strong. This composition can be described as interconnected matrices of carbon and ceramic components. As the effect of increasing the holding time on carbon matrix is irrelevant, the only effect is due to ceramic matrix strengthening.

iv) In Figure 5, they are also included the results corresponding to SiC/CNFs (80/20) vol.% nanocomposite processed by attrition milling. In the previous section it was shown how this processing method improves the material homogeneity and therefore, it allows obtaining materials with higher density and mechanical strength. Thanks to the combined effect of most efficient processing method (attrition milling) and increased holding time (30 min) it

is possible to prepare a SiC/CNFs nanocomposite with a fracture strength >300 MPa.

In summary, there is a positive interaction between SiC and CNFs components. Carbon nanofibers have an excellent potential as functional additive, acting as sintering aid of silicon carbide and functional component of the obtained nanocomposite.

3.4. Composites obtained by sol-gel method

In previous sections, different parameters related with sintering cycle design (maximum temperature or holding time) and material processing method have been considered in order to improve the final properties of the composites. A wide range of material compositions was studied. Although SiC/CNFs (80/20) vol.% is the most promising nanocomposite for structural applications the remaining porosity limits its mechanical performance. In this section, a combination of material modification and the use of optimized sintering cycle will be explored.

A sol-gel synthesis route was studied for the preparation of these nanocomposites. Alumina surface coating on CNFs has been achieved by the procedure published elsewhere [34]. Briefly, in this process it is formed a three dimensional network which perfectly interconnect the sol particles in the whole volume. As carbon nanofibers have been previously dispersed in the sol, they are obtained carbon nanofibers homogeneously coated by the gel layer. Then, heat treatment leads to the formation of alumina nanoparticles. CNFs+10 vol.% Al_2O_3 powders were mixed with silicon carbide nano-powders in order to obtain a SiC/CNFs-Al_2O_3 (80/18-2) vol.% nanocomposite by the two processing methods (US and AM). Figure 6 shows the relative density and fracture strength of the materials obtained. The properties of SiC/CNFs (80/20) vol.% composite are included as reference.

The addition of 2 vol.% alumina clearly enhances nanocomposites densification as it is showed in the fracture surface SEM micrograph of AM SiC/CNFs (80/20) vol.% composite (Figure 7). Materials density after 1 min sintering at 1800 ºC is around 90% t.d while the sol gel modification allows obtaining fully dense composites at 1800 ºC and 30 min holding time. Then, from the kinetic point of view it is necessary to exceed a threshold time in order to remove completely the residual porosity. As consequence, mechanical properties were markedly improved, obtaining nanocomposites with fracture strength ~600 MPa.

Concerning processing method, attrition milling processed composites shows better mechanical properties while the densities are similar to ultrasonic dispersed samples. It can be expected a more efficient mixture of SiC and CNFs-Al_2O_3 components by attrition milling. The fracture strength improvement is related with a stronger interaction between components. As the sintering additive is incorporated on CNFs surface, if the mixture is more homogeneous, the efficiency of alumina acting as sintering aid will be improved. Thus, the nanocomposites obtained by the attrition milling of a mixture of the surface coating of CNFs with an alumina precursor and SiC commercial powders leads to considerably higher fracture strength than material processing by mixed powders.

Then, dense CNFs-reinforced SiC nanocomposites were prepared through combination of control of sintering parameters, selection of suitable processing method and chemical functionalization of carbon nanofibers surface. In the case of SiC/CNFs (80/20) vol.% composite, it is remarkable that besides its high mechanical strength (~600 MPa), this material has additional functionalities that make it interesting for different industrial

application. Thus, its electrical resistivity is as low as 2 10^{-1} Ω cm allowing the application of electro discharge machining processes for obtaining complex shape components based on this material, and a weight reduction of 10% in comparison with monolithic SiC materials.

4. CONCLUSIONS

CNFs/SiC nanocomposites were prepared by different processing routes and sintered by PECPS at different temperatures and holding times. In this study, the addition of CNFs in nanophase SiC matrix was investigated based on densification, microstructure and mechanical strength.

The use of attrition milling as processing method allows obtaining dense CNFs/SiC nanocomposites with very promising properties. The effective reduction of agglomerates leads to superior fracture strength when compared with ultrasonication processed materials. Moreover, carbon nanofilaments features are preserved after milling process reducing the electrical resistivity of composite to 2 10^{-1} Ω cm.

The surface coating of carbon nanofibers using an alumina precursor is a very interesting method to obtain dense CNFs/SiC nanocomposites by enhancing interaction between ceramic and carbon phase. The combination of sol-gel surface modification, high-energy attrition milling of SiC+20 vol.% CNFs powders and pulsed electric-current pressure sintering technique allows obtaining a nanocomposite that combines improved mechanical strength and electrical conductivity and weight reduction.

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References

[1] A.E. McHale (Ed.), Fig. 8666 in Phase Equilibria Diagrams, Phase Diagrams for Ceramists, vol. X, Borides, Carbides, and Nitrides, American Ceramic Society, Columbus, OH, 1994.

[2] S. Prochazka, R.M. Scanlan, J. Am. Ceram. Soc. 58 (1975) 72-73.

[3] H. Endo, M. Ueki, H. Kubo, J. Mater. Sci. 26 (1991) 3769-3774.

[4] M. Singh, D.R. Behrendt, J. Mater. Sci. Eng. A 194 (1995) 193-200.

[5] S. Iijima, Nature (1991) 354-356.

[6] M. Endo, Y.A. Kim, T. Hayashi, K. Nishimura, T. Matushita, K. Miyashita, M.S. Dresselhaus, Carbon 39 (2001) 1287-1297.

[7] A. Merkoci, Microchim Acta 152 (2006) 157-174.

[8] K. Hirota, Y. Takaura, M. Kato, Y. Miyamoto, J. Mater. Sci. 42 (2007) 4792-4800.

[9] S. Maensiri, P. Laokul, J. Klinkaewnarong, V. Amornkitbamrung, Mater. Sci. Eng. A 447 (2007) 44-50.

[10] P. Hvizdoš, V. Puchý, A. Duszová, J. Dusza, Scripta Mater. 63 (2010) 254-257.

[11] Cs. Balazsi, Z. Kónya, F. Wéber, L.P. Biró, P. Arató, Mater. Sci. Eng. C 23 (2003) 1133-1137.

[12] J. Tatami, T. Katashima, K. Komeya, T. Meguro, T. Wakihara, J. Am. Ceram. Soc. 88 (2005) 2889-2893.

[13] J.W. An, D.H. You, D.S. Lim, Wear 255 (2003) 677-681.

[14] R.W. Siegel, S.K. Chang, B.J. Ash, J. Stone, P.M. Ajayan, R.W. Doremus, LS. Schqdler, Scripta Mater. 44 (2001) 2061-2064.

[15] Ch. Laurent, A. Peigney, O. Dumortier, A Rousset, J. Eur. Ceram. Soc. 18 (1998) 2005-2013.

[16] J. Sun, L. Gao, Carbon 41 (2003) 1063-1068.

[17] R.G. Joanna, Z. Antonios, Mater. Sci. Eng. A 287 (2000) 171-177.

[18] W. Yucheng, F. Zhengyi, Mater. Sci. Eng. B 90 (2002) 34-37.

[19] N. Tamari, T. Tanaka, K. Tanaka, I. Kondoh, M. Kawahara, M. Tokita, J. Ceram. Soc. Jpn. 103 (1995) 740-742.

[20] K. Shimoda, T. Hinoki, A. Kohyama, Comp. Sci. Technol. 70 (2010) 387-392.

[21] K. Hirota, H. Hara, M. Kato, Mater. Sci. Eng. A 458 (2007) 216-225.

[22] X. Wang, N.P. Padture, H. Tanaka, Nat. Mater. 3 (2004) 539-544.

[23] L. Ci, J. Wei, B. Wei, J. Liang, C. Xu, D. Wu, Carbon 39 (2001) 329-335.

[24] B. Bittmann, F. Haupert, A.K. Schlarb, Ultrason. Sonochem. 16 (2009) 622-628.

[25] A.F. Kirstein, R.M. Woolley, J. Res. Nat. Bur. Stand. C 71 (1967) 1-10.

[26] F.F. Vitman, V.P. Pukh, Zavod Lab. 29 (1963) 863-867.

[27] ASTM Standard F394-78, STM Annual Book of Standards, Vol. 15.02, Section 16. American Society for Testing and Materials, Philadelphia PA; 1996:466-490.

[28] E. Ermer, P. Wiesław, S. Ludosław, Sol. Stat. Ion. 141-142 (2001) 523-528.

[29] H. Endo, M. Ueki, H. Kubo, J. Mater. Sci. 26 (1991) 3769-3774.

[30] S. Prochazka, P. Popper (Eds.), Special Ceramic, England, vol. 6, 1975, pp. 171-182

[31] C. Greskovich, J.H. Rosolovski, J. Am. Ceram. Soc. 59 (1976) 336-343.

[32] S. Suzuki, H.T. Boron, J. Am. Ceram. Soc. 63 (1980) 349-350.

[33] K.L. More, C.H. Carter, J. Bentley, W.H. Waldin, J. Am. Ceram. Soc. 69 (1986) 695-698.

[34] A. Borrell, V.G. Rocha, R. Torrecillas, A. Fernández, Comp. Sci. Technol. 71 (2011) 18-22.

[35] A. Borrell, V.G. Rocha, R. Torrecillas, A. Fernández, J. Am. Ceram. Soc. 94 (2011) 2048-2052.

[36] A. Borrell, A. Fernández, C. Merino, R. Torrecillas, Int. J. Mater. Res. 101 (2010) 112-118.

[37] O.A. Orumwense, E. Forssberg, Min. Proces. Extrac. Metall. Rev. 11 (1992) 107-127.

[38] H. Persson, E. Forssberg, Aufbereitungs Technik. 35 (1994) 307-319.

[39] A. Iftikhar, C. Hongzhi, C. Huahui, Z. Huiyou, K. Andrew, Q.Z. Yan, J. Eur. Ceram. Soc. 30 (2010) 865-873.

Figure Captions:

Figure 1. Fracture strength of CNFs/SiC nanocomposites as a function of sintering temperature and CNFs content.

Figure 2. SEM fracture surfaces of CNFs+50 vol.% SiC nanocomposites sintered by PECPS at 1600 ºC (a) and 1800 ºC (b).

Figure 3. Comparison of the relative density and fracture strength of the sintered materials after processing by US (ultrasonic dispersion of powder mixtures) and AM (attrition milling of powder mixtures).

Figure 4. SEM Fracture surface of CNFs/SiC nanocomposite with 20 vol.% of CNFs sintered at 1800 ºC-1 min: *left* is ultrasonic dispersion and *right* is attrition milling.

Figure 5. Influence of holding time on densification and fracture strength of CNFs/SiC US mixture powders sintered by PECPS at 1800 ºC. CNFs/SiC (20/80) vol.% density and fracture strength of the attrition milled composite are overwritten with cross markers.

Figure 6. Relative density and fracture strength of CNFs/SiC composites obtained from mixing powders and sol-gel method and fabricated by PECPS at 1800 ºC and 80 MPa for 1 and 30 min in vacuum.

Figure 7. SEM micrograph of AM SiC/CNFs sol-gel (80/20) vol.% composite sintered at 1800 ºC during 30 min.

Figure 1

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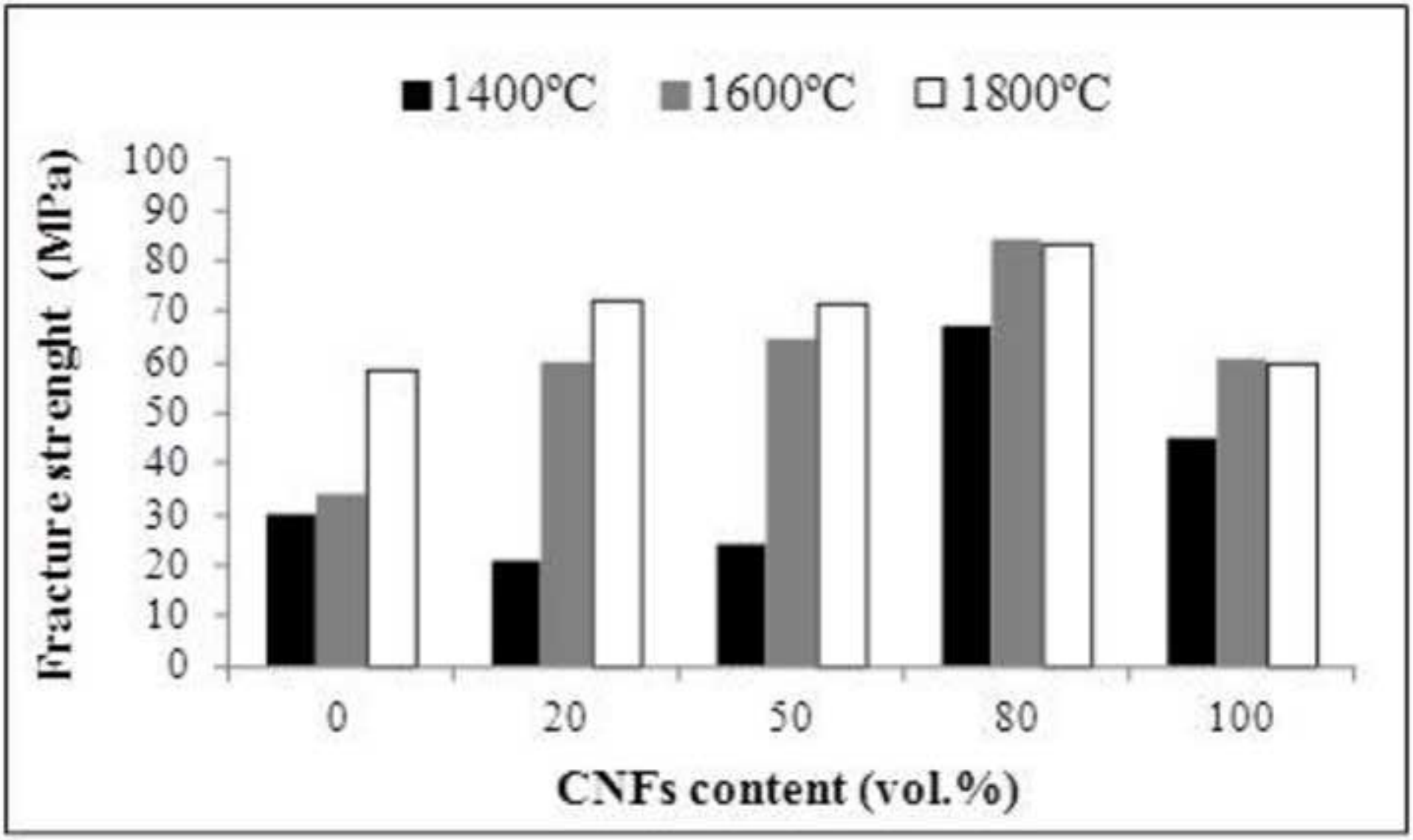

Figure 2

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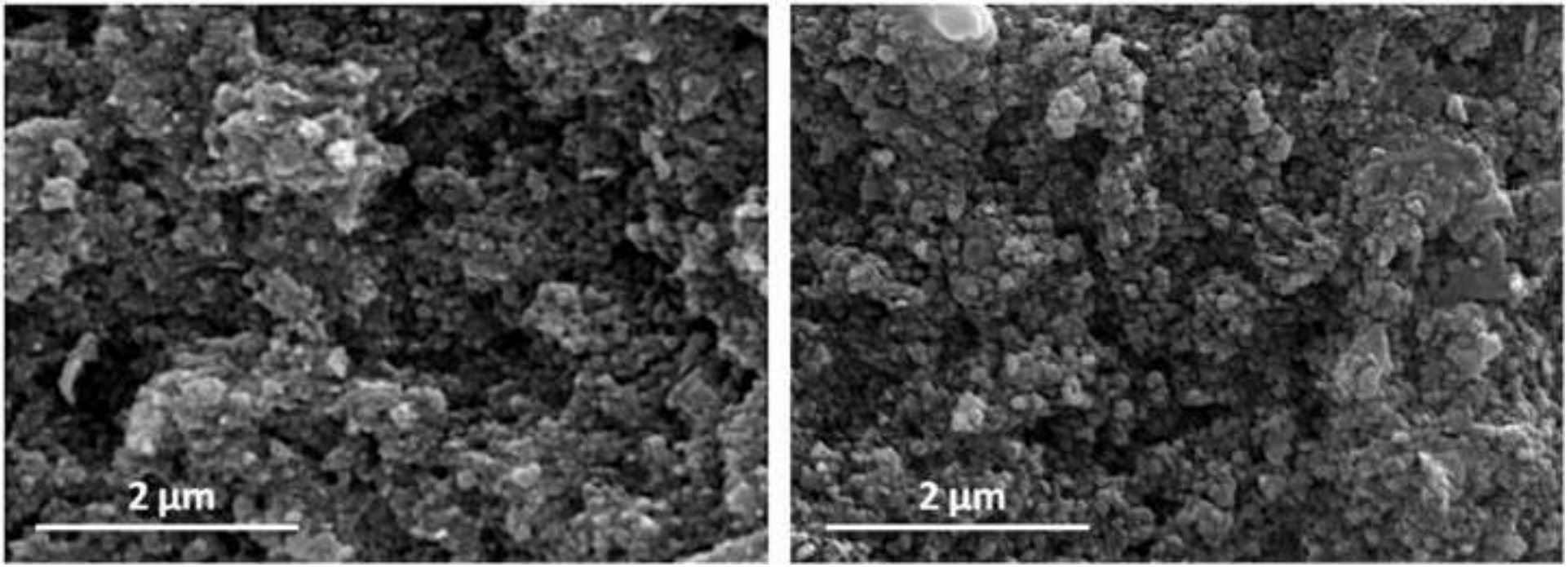

Figure 3

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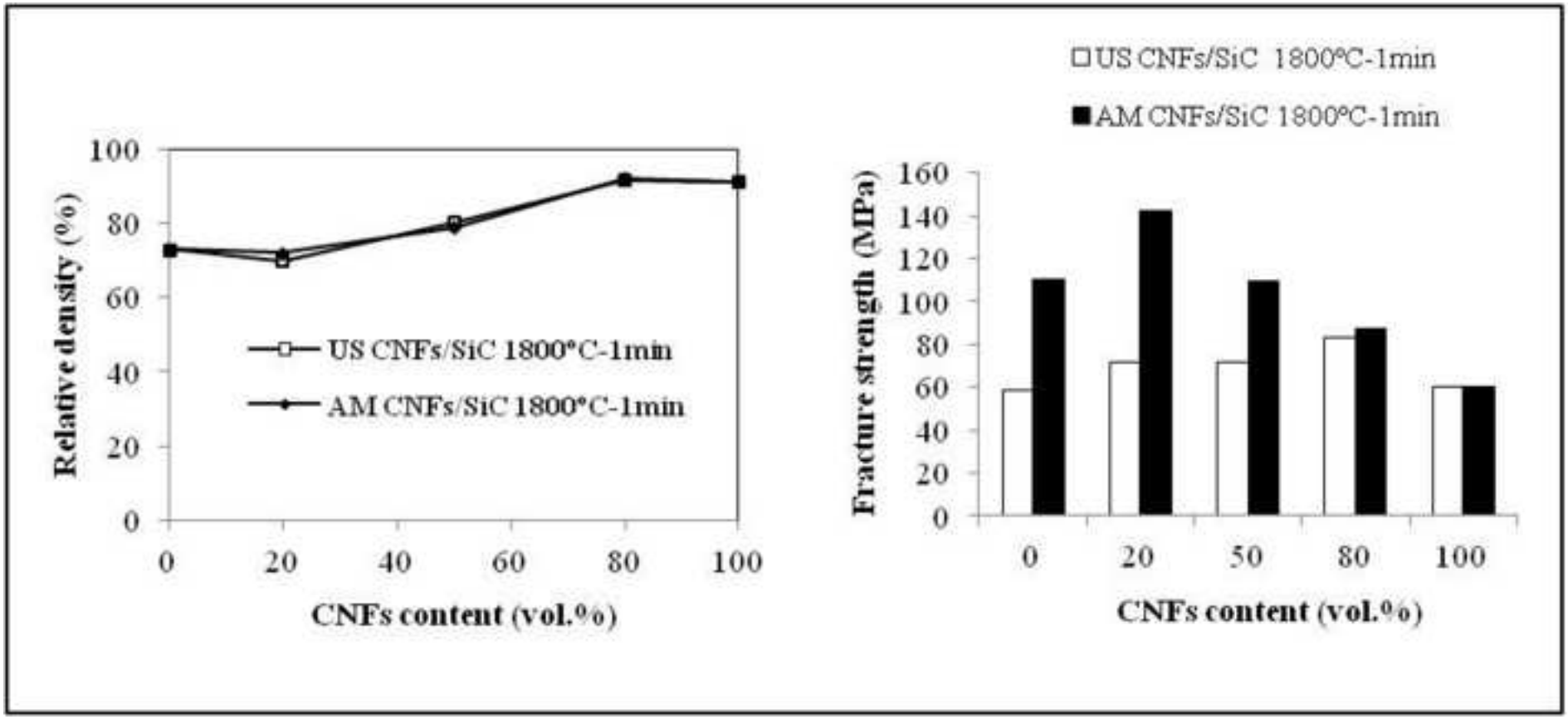

Figure 4

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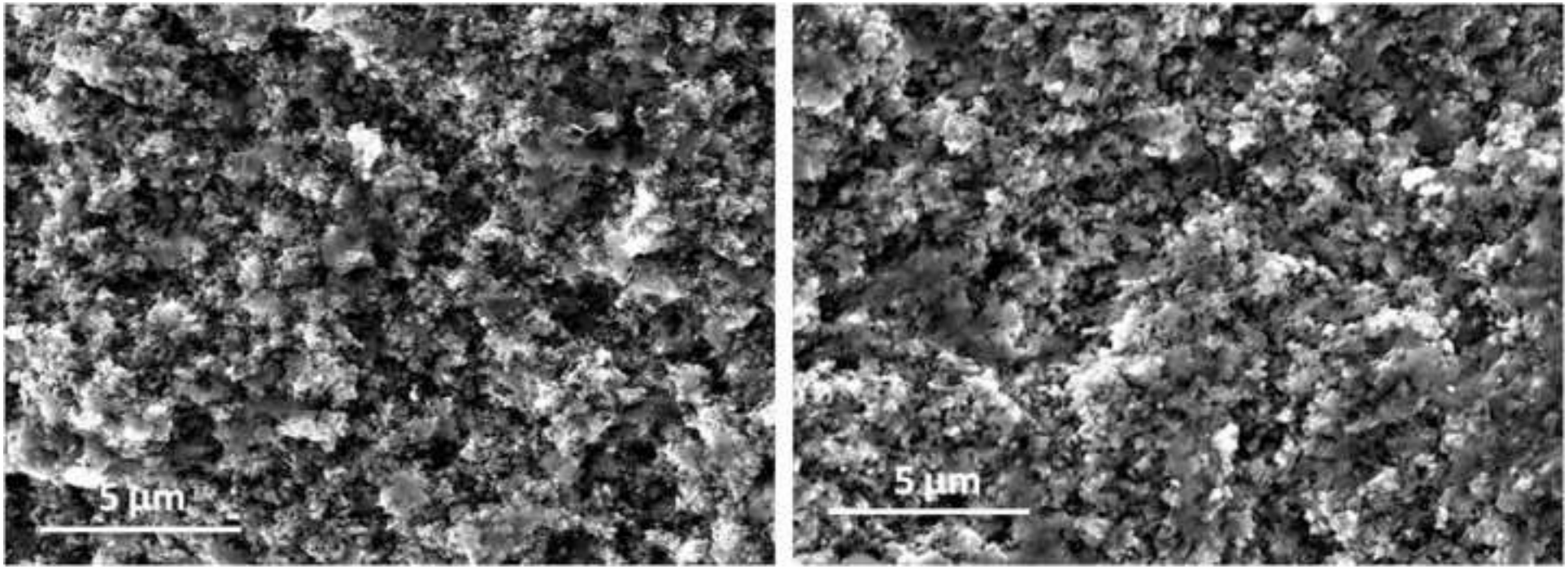

Figure 5

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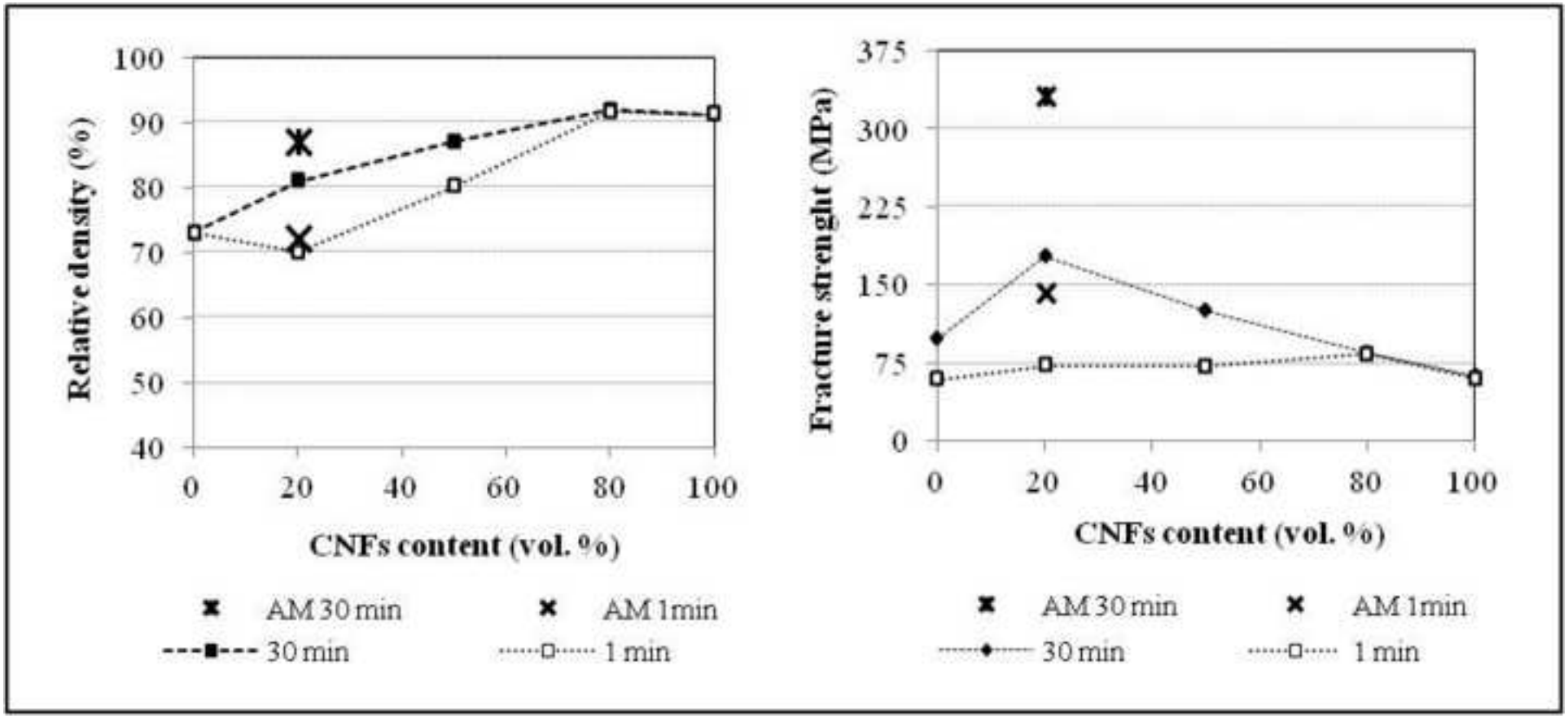

Figure 6

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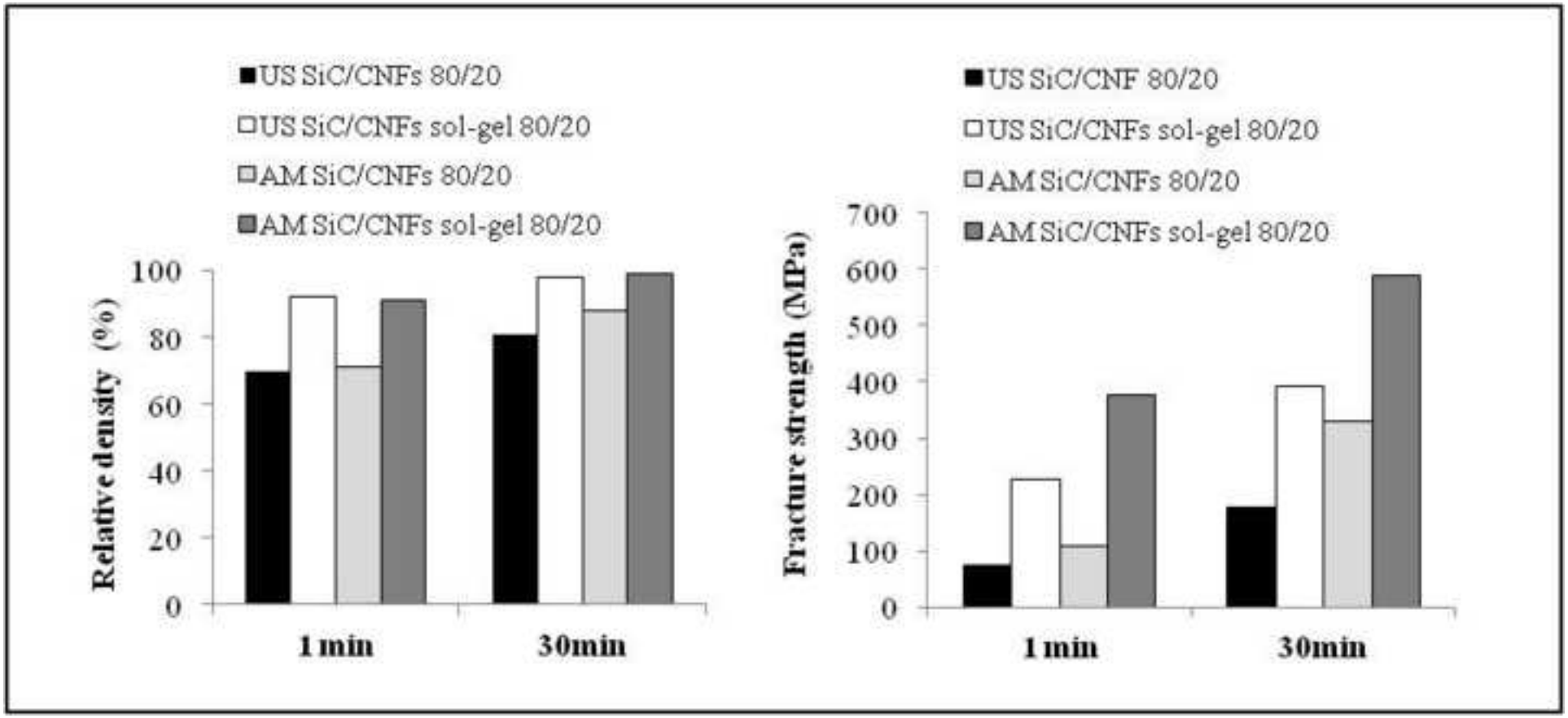

Figure 7

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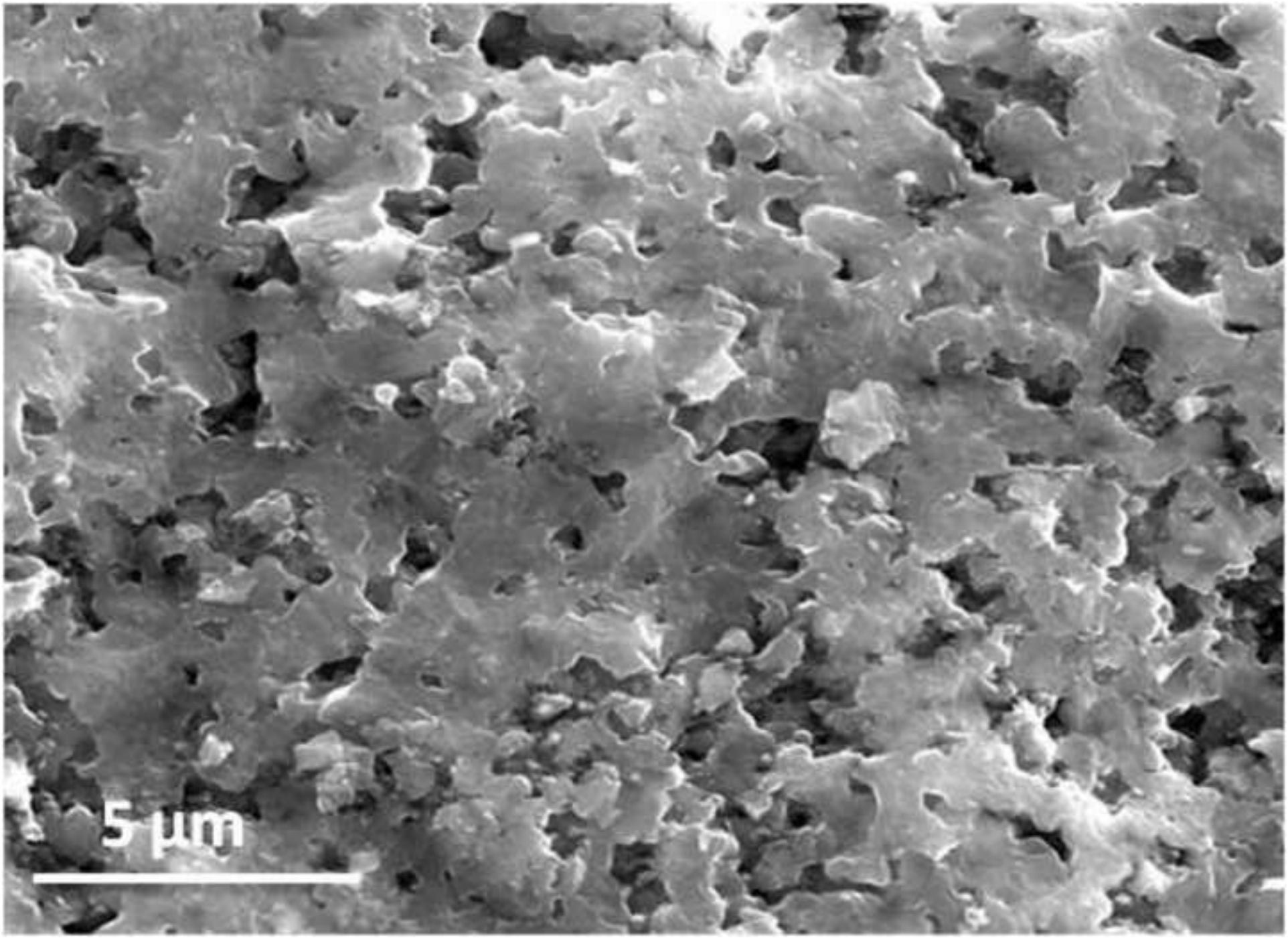

Table 1

%vol. CNFs	1400 ºC	1600 ºC	1800 ºC
	Relative density (%)		
0	58,0	65,2	73,0
20	57,7	61,9	70,0
50	70,1	73,3	80,2
80	86,2	88,1	91,6
100	84,0	91,0	91,2

Table 1. Relative density of monolithic SiC, CNFs material and CNFs/SiC nanocomposites sintered by PECPS at 1400, 1600 and 1800 ºC and 80 MPa for 1 min under vacuum.