

## Synthesis and stability of Ti-C/nano-ceramics composites

Ahmed Al-Ghaban, Niveen Jamal, and Hadeel Ahmed

<sup>1</sup>University of Technology – Baghdad, Materials Engineering Department

Email: [hudhudalbaiaty@yahoo.com](mailto:hudhudalbaiaty@yahoo.com) , [a.ghabban@gmail.com](mailto:a.ghabban@gmail.com)

**Abstract.** In this paper the phase stability of compacted Ti-C/ ceramic nanoadditives composites was investigated. A mixture of elemental Ti and Graphite powders with a range of ceramic nanoadditives of each  $\text{Al}_2\text{O}_3$  and CuO was sintered at 1100 °C; the phase growth was then assessed. X-ray diffraction (XRD) has been used to identify the resultant phases at different nano additives percentage and different various pressures. Differential thermal analysis (DTA) was also used to examine the phase evolution during sintering. The results show that TiC is the most stable phase when no additives have been added.  $\text{TiO}_2$  is found to be the widely prevalent oxide in the case of Ti-C/ nano  $\text{Al}_2\text{O}_3$  and also for Ti-C/ nano CuO mixture. In the case of Ti-C/ nano CuO, it is suggested to have two stage of reaction where  $\text{Cu}_2\text{O}$  appears to be formed first at 650-700 °C.

**Keywords:** phase stability, nanoadditives, Titanium Carbide, ceramic composites

### 1. Introduction

Recently, the focus of many researchers has been directed to nanostructured materials as advanced materials due to their promising physical and mechanical properties (1). Special attention has also been given to nanocomposites since their properties were started to be investigated such as the high strength and toughness as well as reduction of grain size (2). The ceramic-matrix composites are of rising concern since they can promote the low fracture resistance of ceramics (3). One of the most material systems with such advantages are carbides and their composites according to their promising combination of properties (3). Transition metal carbides and nitrides have the potential for use in the most difficult and extreme conditions as due to the spectrum of properties for both ceramic and metallic materials (4). The ceramic material of TiC is one of the transition metal carbide which has many applications in high-tech included mechanical, chemical, and microelectronics (5). From long series of candidates TiC stands to be an effective material due to its high thermal conductivity, high corrosion resistance, high hardness, high melting point, and low neutron absorption cross-section (6). On the other hand, its densification is tricky by classic sintering due to its strong covalent bond (6-8). Further more such systems must be managed under high temperatures according to its high melting point and low diffusion coefficient (9, 10). It is well known that introducing of additives to such ceramic system effects to reduce the sintering temperature (8, 11). Moreover, some ceramic additives suggested to improve the mixture wettability and to get much more dense bulk products or to improve the mechanical properties of the final product (6, 12-15). Other additive such as CuO was reported to reduce the sintering temperature of ceramics (16). In this paper, additives of  $\text{Al}_2\text{O}_3$  and CuO have been mixed up with the base of Ti-C elemental powders to investigate the effect of these nano powders on the final physical and mechanical properties of TiC. The goal of this work is to



contribute along part of this direction studying the phase stability of TiC/nano additives composites upon sintering.

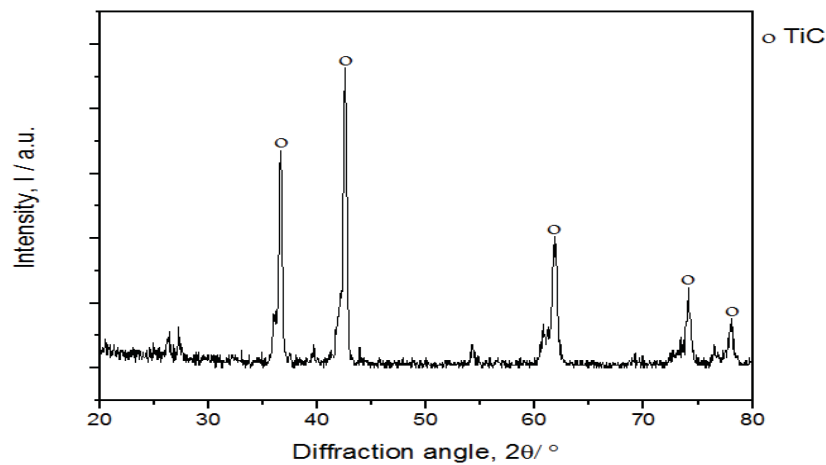
## 2. Experimental

Raw powders of Titanium (>99%, 10  $\mu\text{m}$ ), and Graphite (>99%, 1  $\mu\text{m}$ ) were weighed in a convenient weight percentage ratio of TiC. These were mixed together using mechanical mixer for 2 h. The mixed powder was then mixed as group 1 with 2%, 4%, and 6%  $\text{Al}_2\text{O}_3$ , respectively. The second group was mixed with 2%, 4%, and 6%  $\text{CuO}$ , respectively. The samples from each group were then formed by uniaxial pressing at loads of 5, 10, and 15 ton, respectively. The samples were sintered in an Ar atmosphere at 1100  $^{\circ}\text{C}$  for 1 h. The used heating chamber was programmed to achieve the temperature of 500  $^{\circ}\text{C}$  with the rate of 40  $^{\circ}$  per min and then to 1100  $^{\circ}\text{C}$  with the rate of 20  $^{\circ}$  per min. The samples were then cooled down slowly inside the heating chamber. The phases of the sintered samples were identified by X-ray diffraction analysis (XRD-6000 Shimadzu) employing  $\text{Cu K}\alpha$ , the measuring settings was 60 Kv and 80 mA. The LINSEIS STA Platinum Series (simultaneous thermal analysis) was utilized to investigate the thermal analysis of a sample.

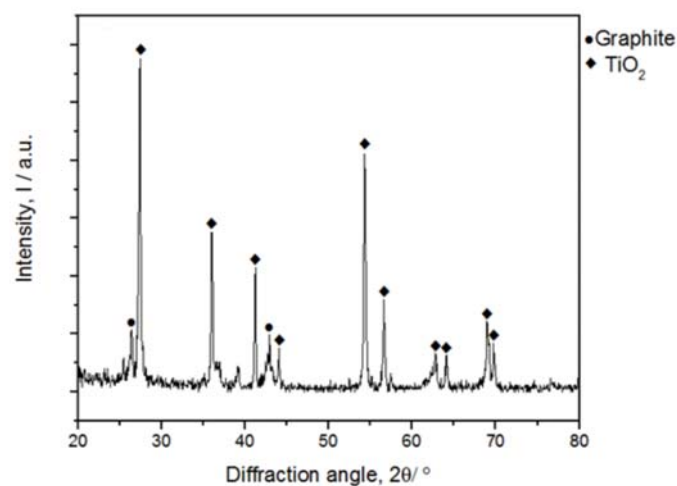
## 3. Results and discussion

The X-ray diffraction pattern of pure Ti-C sample is shown in Fig. 1. This sample sintered at 1100  $^{\circ}\text{C}$  from mixed elemental powders of Titanium and Graphite. The XRD analysis shows that almost all appeared peak matched the cubic phase of TiC. Hence, titanium carbide suggested being the most stable phase along the procedure mentioned in this paper due to the direct contact between Ti and C. The temperature range presented here is much less compared with what presented before for many researchers (6, 7, and 9). Adding nano additives to the mixture of Ti-C, many changes on the properties of the final product would be expected. Therefore, the results obtained from this research will be in the interest of the trend to compare the final properties with the base material. The focus will be on the stability of the resulting phases of this transaction. Fig. 2 shows the XRD result of 1100  $^{\circ}\text{C}$  sintered 2%  $\text{Al}_2\text{O}_3$  doped in Ti-C composite ceramic. Almost all appeared peaks match the structure of  $\text{TiO}_2$ . With further increasing of  $\text{Al}_2\text{O}_3$  doped in Ti-C mixture up to 6%, the  $\text{TiO}_2$  structure is the most dominant phase as shown in fig. 3. Many articles have dealt with this subject, none of which has indicated the stability of the resulting phases during the process of direct interaction (17-19). Mehrizi obtained the formation of  $\text{TiO}_2$  during the ball-milling process to form  $(\text{TiC}/\text{Al}_2\text{O}_3)$  nanocomposite (20). It is good to know that the resultant  $\text{TiO}_2$  is obtained at all pressure used. Fig. 4 shows the XRD result of 4 %  $\text{CuO}$  doped in Ti-C composite ceramic sintered at 1100  $^{\circ}\text{C}$ . The results obtained in this case are very similar to those of nano-alumina addition. There are not many literatures mentioned about the interaction of  $(\text{Ti-C}/\text{CuO})$  nanocomposites. In the DTA process, the result of DTA of 1100  $^{\circ}\text{C}$  sintered 2%  $\text{Al}_2\text{O}_3$  doped in Ti-C composite ceramic is shown in fig. 5. This result suggests one direct contact reaction to form  $\text{TiO}_2$  due to one exothermal peak in the temperature range of 660-770  $^{\circ}\text{C}$ . No further reaction could be observed and the rest of residual Al may ingress inside the  $\text{TiO}_2$  structure at the Ti positions. The same results were obtained for the rest  $\text{Al}_2\text{O}_3$  concentrations of 4 % and 6 %, respectively. Hence, no additional subsidence reactions would be expected due to increased  $\text{Al}_2\text{O}_3$  concentration. For the purpose of making a logical comparison between the two different nano additions, DTA was also used to investigate the thermal stability of  $(\text{Ti-C}/\text{CuO})$  nanocomposite. Figure 6 shows the result of DTA of 1100  $^{\circ}\text{C}$  sintered 2%  $\text{CuO}$  doped in Ti-C composite. Here, two overlapped exothermal peaks in the temperature range of 630-730  $^{\circ}\text{C}$  were observed. As compared with 2%  $\text{Al}_2\text{O}_3$  doped in Ti-C composite, the range of reaction temperature is observed to be reduced. In order to investigate the early presented product according to DTA results, XRD was utilized to identify that product during sintering at the temperature range of about 700  $^{\circ}\text{C}$ . Figure 7 presents the XRD result of 2%  $\text{CuO}$  doped in Ti-C composite after sintering at 700  $^{\circ}\text{C}$  as compared with the XRD result of the final  $\text{TiO}_2$  product of the same material system. In a closer look, it can be said that the reaction involves the production  $\text{Cu}_2\text{O}$  first then this will be converted to  $\text{TiO}_2$  at the second stage. Pike et al.

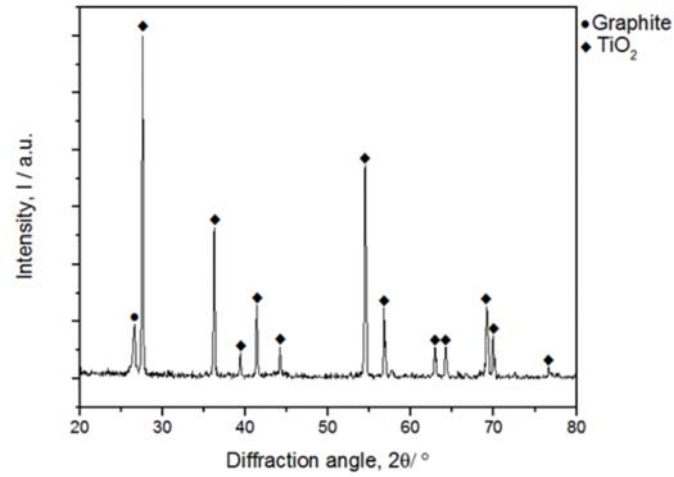
mentioned the reduction of CuO nanoparticles to form the stable Cu<sub>2</sub>O (21). Goldstein et al. gave the sequence of copper oxide reduction to Cu<sub>2</sub>O and then to Cu with the presence of carbon monoxide (22). It is logical to believe that there is great agreement with the results reviewed in this research and what has been mentioned in previous literature.



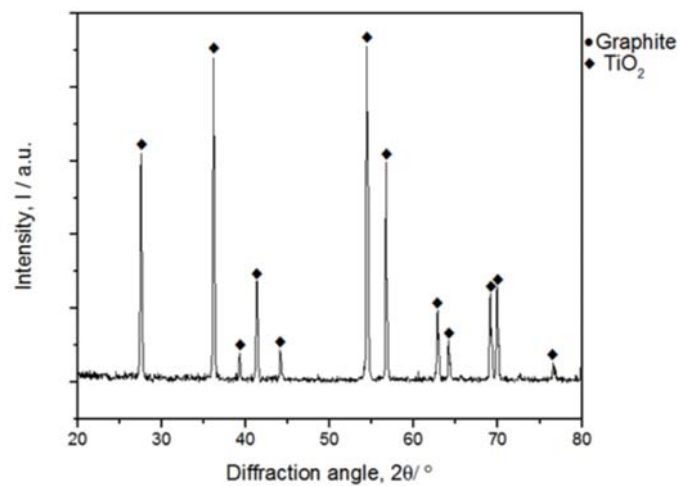
**Figure 1:** The XRD result of Ti-C mixture sintered at 1100 °C



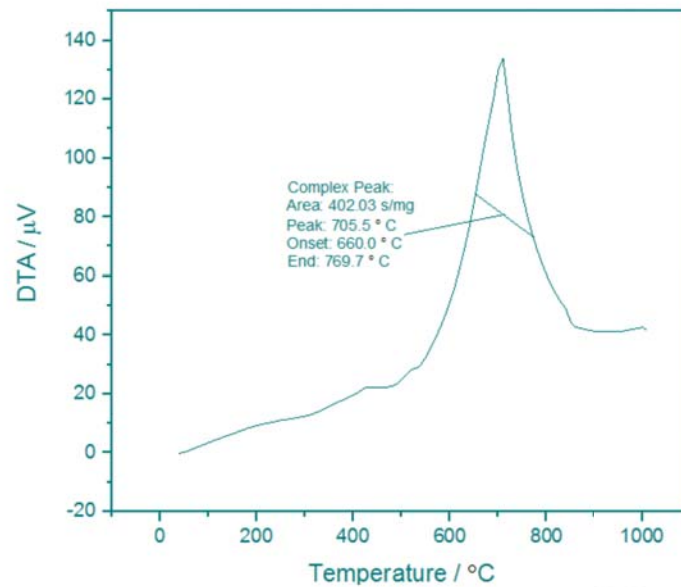
**Figure 2:** The XRD result of 1100 °C sintered sample with 2 % Al<sub>2</sub>O<sub>3</sub> doped in Ti-C composite



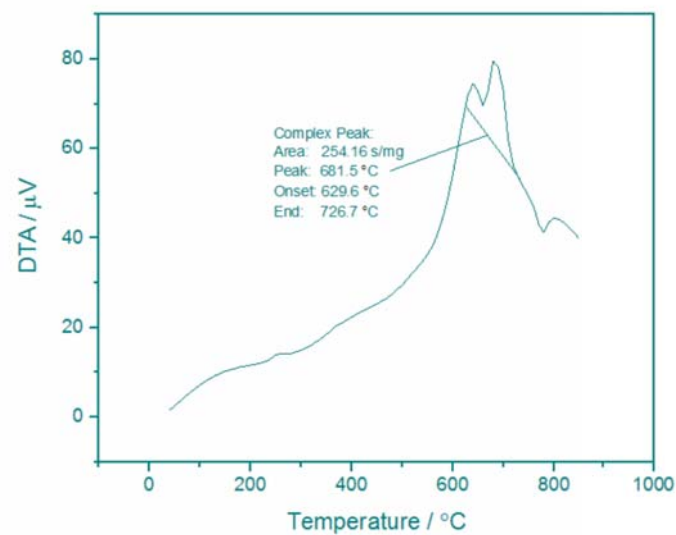
**Figure 3:** The XRD result of 1100 °C sintered sample with 6 % Al<sub>2</sub>O<sub>3</sub> doped in Ti-C composite



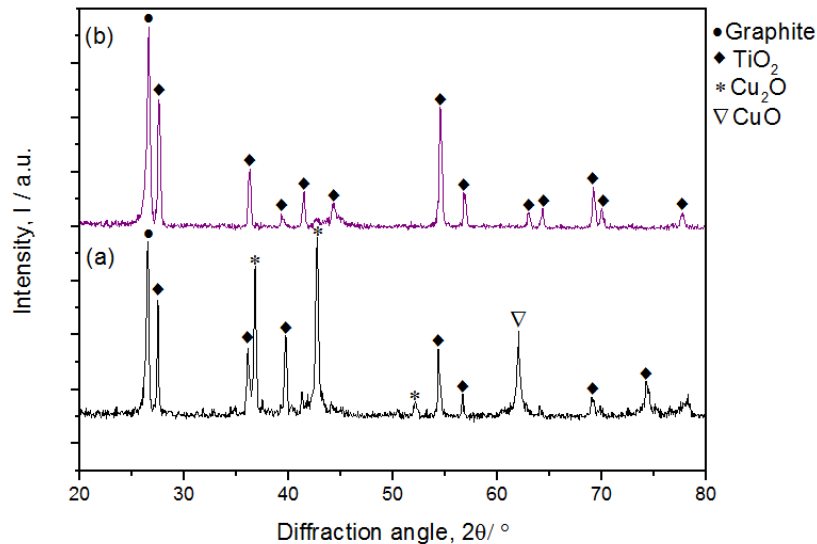
**Figure 4:** The XRD result of 1100 °C sintered sample with 4 % CuO doped in Ti-C composite



**Figure 5:** The DTA result of 1100 °C sintered sample with 2 %  $\text{Al}_2\text{O}_3$  doped in Ti-C composite



**Figure 6:** The DTA result of 1100 °C sintered sample with 2 %  $\text{CuO}$  doped in Ti-C composite



**Figure 7:** The XRD results of: (a) Ti-C with 2 % CuO nanocomposite sintered at 700 °C (b) Ti-C with 2 % CuO nanocomposite sintered at 1100 °C

#### 4. Conclusions

The following conclusions can be set from the results presented in this paper

- 1- At 1100 °C the cubic TiC phase found to be stable from the direct reaction between Titanium and Graphite.
- 2- Adding nano additives of Al<sub>2</sub>O<sub>3</sub> and CuO to Ti-C substance helps to stabilize the TiO<sub>2</sub> phase after sintering at 1100 °C.
- 3- TiO<sub>2</sub> forms to be the most stable phase at 1100 °C with one step direct reaction between Ti-C and nano Al<sub>2</sub>O<sub>3</sub>.
- 4- TiO<sub>2</sub> forms to be the most stable phase at 1100 °C with two steps indirect reaction between Ti-C and nano CuO where Cu<sub>2</sub>O forms first at moderate temperatures then it is reduced to form the final product.

#### References

1. Abderrazak H, Schoenstein F, Abdellaoui M and Jouini N 2011 *Int. Journal of Refractory Metals and Hard Materials* **29** 170.
2. Nayak P, Lin H., Chang M, Chen W and Huang J 2013 *Journal of the European Ceramic Society* **33** 3095.
3. Vallauri D, Adri'an I and Chrysanthou A 2008 *Journal of the European Ceramic Society* **28** 1697.
4. Baillet J, Gavarini S, Millard N, Garnier V, Peaucelle C, Jaurand X, Cardinal S, Duranti A, Bernard C, Rapegno R, Dernoncourt L and De Echave T 2016 *Journal of the European Ceramic Society* **36** 3099.

5. Koc R and Folmer J 1997 *Journal of the American Ceramic Society* **80** 952.
6. Cheng L, Xie Z, Liu G, Liu W and Xue W 2012 *Journal of the European Ceramic Society* **32** 3399.
7. Xue J, Liu J, Zhang G, Zhang H, Liu T, Zhou X and Peng S 2016 *Scripta Materialia* **114** 5.
8. Cheng L, Xie Z and Liu G 2013 *Journal of the European Ceramic Society* **33** 2971.
9. Fu Z and Koc R 2017 *Ceramics International* **43** 17233.
10. Zhang X, Hilmas G and Fahrenholtz W 2007 *Journal of the American Ceramic Society* **90** 393.
11. Fu Z and Koc R 2016 *Materials Science & Engineering* **676** 278.
12. Park S and Kang S. 2005 *Scripta Materialia* **52** 129.
13. Park C, Nam S and Kang S 2016 *Materials Science & Engineering* **649** 400.
14. Cabrero J, Audubert F and Pailler R 2011 *Journal of the European Ceramic Society* **31** 313.
15. Liu N, Xu Y D, Li H, Li G H and Zhang L D 2002 *Journal of the European Ceramic Society* **22** 2409.
16. Kim J H, Kim J S, Han S H, Kang H W, Lee H G and Cheon C I 2017 *Materials Research Bulletin* **96** 121.
17. Fei Y H, Huang C Z, Liu H L and Zou B 2014 *Ceramics International* **40** 10205.
18. Cai K F, McLachlan D S, Axen N and Manyatsa R 2002 *Ceramics International* **28** 217.
19. Cheng Y, Sun S and Hu H 2014 *Ceramics International* **40** 16761.
20. Mehrizi M Z, Beygi R, Mostaan H, Raoufi M and Barati A 2016 *Ceramics International* **42** 17089.
21. Pike J, Chan S W, Zhang F, Wang X and Hanson J 2006 *Applied Catalysis A: General* **303** 273.
22. Goldstein E A and Mitchell R E 2011 *Proceedings of the Combustion Institute* **33** 2803.