

Effect of Y_2O_3 addition on the densification and mechanical properties of alumina–niobium carbide composites

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Abstract

Alumina-based composites reinforced with refractory carbides are potential cutting tool materials. They exceed the capabilities of cemented carbides with respect to hot hardness and thermal stability, resulting in faster cutting speeds. Liquid-phase sintering of Al_2O_3 –NbC composites was investigated as an alternative to pressure-assisted processes. Al_2O_3 reinforced by NbC (5–40 wt.%) was sintered with 3 wt.% Y_2O_3 . In order to assess the effect of the formation of a liquid phase on the properties of the composites, sintering was carried out either below or above the Al_2O_3 – Y_2O_3 eutectic temperature, at 1650 and 1800°C, respectively. Density, hardness, fracture toughness and wear resistance of the composite materials were evaluated. Liquid phase sintering did not affect the fracture toughness, but improved both the density and the hardness of the material, regardless of its NbC contents. Higher concentrations of NbC increased the wear resistance of the composite. © 2001 Published by Elsevier Science Ltd and Techna S.r.l. All rights reserved.

Keywords: A. Sintering; C. Hardness; D. Al_2O_3 ; E. Wear parts; Niobium carbide

1. Introduction

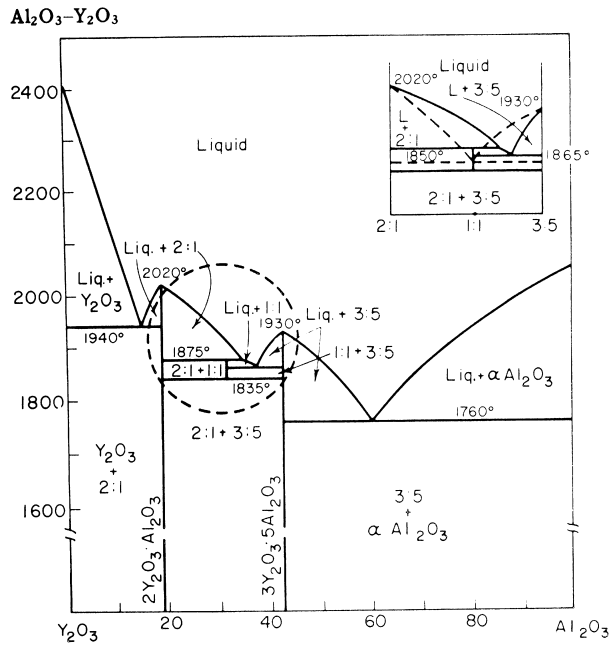
Monolithic ceramics and ceramic composites have been developed as alternative materials for cutting tools, as they resist higher temperatures and can cut faster than cemented carbides [1–5]. Alumina and silicon nitride have been successfully applied to machining a variety of engineering metals and alloys, particularly cast iron, hardened steel and heat-resistant alloys [6]. Its lower coefficient of thermal expansion renders Si_3N_4 excellent thermal shock behaviour, nonetheless Al_2O_3 has a higher chemical stability and wear resistance. Attempts have been made to improve hardness and fracture toughness of alumina, including the production of alumina-based composites reinforced by refractory carbides, especially TiC [6]. The addition of TiC to alumina increases hot hardness, thermal conductivity and

resistance to attrition wear, making the material suitable for fine machining of cast iron.

Niobium carbide can also be used to reinforce alumina, due to its high melting temperature (3600°C), high stiffness (340 GPa) and high hardness (> 20 GPa). Contrary to TiC, TaC and ZrC, NbC maintains high hardness values at temperatures above 500°C, which is an additional advantage for high-speed cutting [7]. However, there have been indications that the hardness of Al_2O_3 –composites reinforced by refractory carbides depend not only on the carbide contents but also on the density of the composite [3,8]. The densification of ceramic matrix composites requires either pressure-assisted or liquid-phase sintering. Hot-pressing and hot-chamber pressing are costly batch processes. On the other hand, small amounts of Y_2O_3 have been successfully used to densify Al_2O_3 –TiC [9] and Al_2O_3 –NbC composites [10]. The binary Al_2O_3 – Y_2O_3 phase diagram (Fig. 1) shows an eutectic for ~24 mol.% Y_2O_3 at 1760°C, and the formation of liquid phase for compositions containing up to ~38 mol.% Y_2O_3 [11]. Therefore the addition of

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Fig. 1. Al_2O_3 – Y_2O_3 phase diagram [12].

small amounts of Y_2O_3 and sintering temperatures higher than 1760°C promote the formation of a liquid phase that assists densification. The effect of the presence of a residual grain-boundary phase on the hardness and fracture toughness of Al_2O_3 –NbC composites should then be investigated.

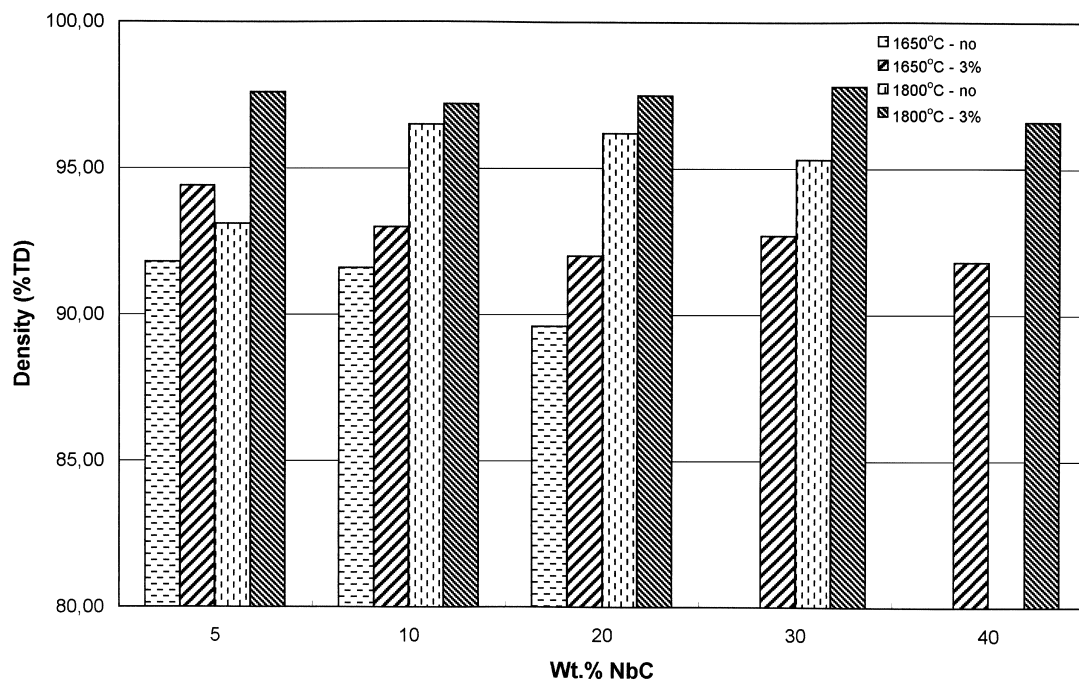
Table 1

Properties of Al_2O_3 –NbC composites sintered with 3 wt.% Y_2O_3

Composition (wt.%)	Density (% TD)	H_v (GPa)	K_{IC} ($\text{Mpa m}^{1/2}$)
<i>Sintering at $1650^\circ\text{C}/30\text{ min}$</i>			
Al_2O_3 –5% NbC	94.4	10.5 ± 1.1	3.8 ± 0.6
Al_2O_3 –10% NbC	93.0	10.7 ± 1.1	3.9 ± 0.7
Al_2O_3 –20% NbC	92.0	12.1 ± 0.6	4.2 ± 0.4
Al_2O_3 –30% NbC	92.7	12.1 ± 2.1	3.2 ± 0.6
Al_2O_3 –40% NbC	91.8	12.0 ± 0.5	3.3 ± 0.9
<i>Sintering at $1800^\circ\text{C}/15\text{ min}$</i>			
Al_2O_3 –5% NbC	97.6	15.5 ± 0.7	2.9 ± 0.4
Al_2O_3 –10% NbC	97.2	16.4 ± 1.6	2.8 ± 0.4
Al_2O_3 –20% NbC	97.5	16.7 ± 1.3	3.6 ± 0.5
Al_2O_3 –30% NbC	97.8	16.9 ± 1.5	3.6 ± 0.5
Al_2O_3 –40% NbC	96.6	16.8 ± 1.2	3.7 ± 0.6

2. Experimental procedure

Alumina APC-2011 SG (Alcoa, Brazil), $D_{50} = 2.3\text{ }\mu\text{m}$, surface area $1.5\text{ m}^2/\text{g}$ and NbC (Hermann Starck Berlin, Germany), $D_{50} = 2.3\text{ }\mu\text{m}$, were mixed with 3 wt.% Y_2O_3 resulting in composites with NbC contents varying from 5 to 40 wt.%. The powder mixtures were milled for 4 h in a planetary mill containing Al_2O_3 grinding balls. Pellets 8 mm in diameter were then uniaxially pressed under 100 MPa and pressureless sintered under flowing argon at $1650^\circ\text{C}/30\text{ min}$ and $1800^\circ\text{C}/15\text{ min}$ in a graphite-resistance-heated furnace. The heating and cooling rates were $20^\circ\text{C}/\text{min}$. The density of the sintered samples was

Fig. 2. Density of Al_2O_3 –NbC composites sintered at 1650 and 1800°C with and without Y_2O_3 .

determined using the Archimedes method. Sintered specimens were polished with diamond paste to a 1 μm finish, and thermally etched under vacuum at 1500–1550°C/30 min. X-ray diffraction analysis was carried out in order to identify whether or not crystalline grain-boundary phases formed upon sintering. Microhardness and fracture toughness were obtained measuring the lengths of the cracks and the diagonal impressed by a Vickers indenter applying loads that varied from 30 to 100 N for 15 s. Loads varied according to the carbide contents of the test specimen in order to produce radial cracks without ramification. The wear resistance of the composites was investigated under dry conditions on a Plint & Partners TE67 pin-on-disk tribometer. A AISI 1045 steel disk with $R_a = 0.67 \mu\text{m}$ was used. The composite pin specimens were fixed against the disk under a load of 70 N. Tests were carried out at a sliding speed of 1.0 m/s. The results were reported as a volume of removed material from both the sample and the testing disk after 2 h.

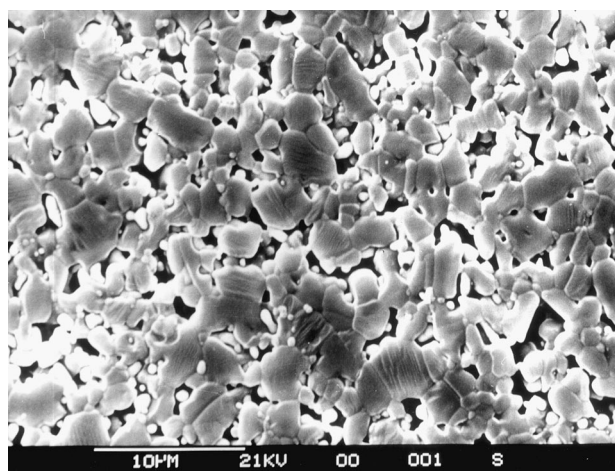


Fig. 3. SEM image of Al_2O_3 -20 wt.% NbC-3 wt.% Y_2O_3 sintered at 1650°C/30 min.

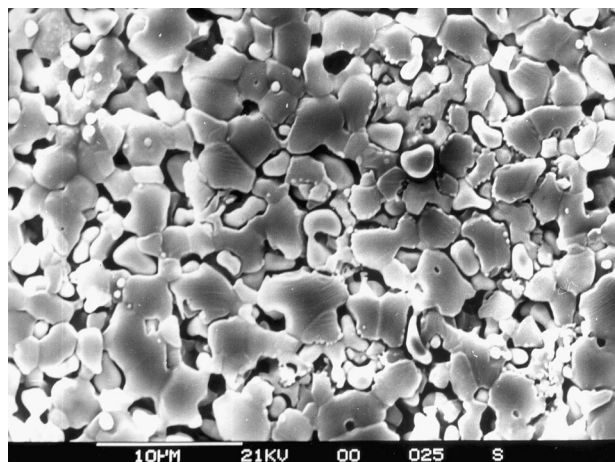


Fig. 4. SEM image of Al_2O_3 -30 wt.% NbC-3 wt.% Y_2O_3 sintered at 1800°C/15 min.

3. Results and discussion

Evaluated parameters of Al_2O_3 -NbC composites sintered with 3 wt.% Y_2O_3 are summarized in Table 1. It can be readily observed that raising the sintering temperature from 1650 to 1800°C improved densification. This is probably related to the formation of a liquid phase from the reaction of Al_2O_3 and Y_2O_3 [11]. Specimens sintered at 1650°C depicted densities ranging from ~92 to 94.5% TD. Sintering at a higher temperature increased density values to ~97–98% TD. It can also be seen from Table 1 that increasing the NbC contents of samples sintered at 1650°C reduced the density of the composite material, which can be attributed to a pinning effect. The presence of dispersed second-phase particles reduces the mobility of alumina grain boundaries during sintering and prevents full densification. Similar behaviour has been observed for a variety of composite systems, both metallic [12] and ceramic [13–15]. Conversely, the density of specimens sintered at 1800°C remained roughly unaffected by the NbC contents. This is probably related to the presence of a liquid phase which increased the mobility of grain boundaries and attenuated the pinning effect. The evolution of the densification of Al_2O_3 -NbC sintered with and without Y_2O_3 is shown in Fig. 2, as a function of both temperature and NbC contents. The effect of the formation of a liquid phase can be determined observing the density of specimens sintered at 1800°C with and without Y_2O_3 (Table 1). For NbC contents up to 30 wt.%, liquid-phase sintering at 1800°C resulted in the highest measured densities. At 1650°C, no liquid phase was formed even in samples containing Y_2O_3 . Consequently, the presence of the additive did not show any significant improvement in the densification of composites with up to 20 wt.% NbC.

Figs. 3 and 4 show the microstructure of Al_2O_3 -NbC-3 wt.% Y_2O_3 composites sintered at 1650°C/30 min and 1800°C/15 min, respectively. Grain growth is quite evident

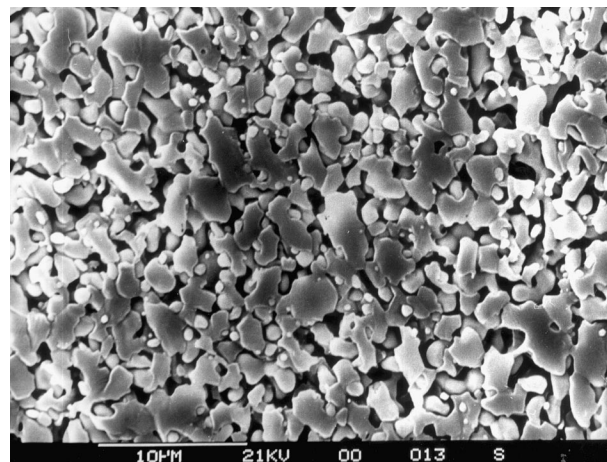


Fig. 5. SEM image of Al_2O_3 -30 wt.% NbC without Y_2O_3 sintered at 1800°C/15 min.

and it can be attributed both to a higher sintering temperature and the formation of liquid phase. The individual influence of the liquid phase on grain growth can be visualised comparing the microstructures of Al_2O_3 –30 wt.% NbC composites sintered at 1800°C , with and without Y_2O_3 (Figs. 4 and 5, respectively). The presence of a liquid phase enhanced grain growth and inhibited the pinning effect of the dispersed particles. X-ray diffraction was carried out for Al_2O_3 –40 wt.% NbC–3 wt.% Y_2O_3 sintered at $1650^\circ\text{C}/30$ min and $1800^\circ\text{C}/$

15 min (Fig. 6). Both specimens revealed the presence of Al_2O_3 and NbC in addition to a crystalline phase of composition $3\text{Y}_2\text{O}_3\cdot 5\text{Al}_2\text{O}_3$ (YAG). Such a phase has a garnet-type structure and is known to be stable under equilibrium conditions [16]. The formation of YAG in Al_2O_3 –NbC– Y_2O_3 composites has been reported for samples sintered at temperatures in the vicinity of the eutectic [11]. However, the fact that diffraction peaks corresponding to YAG were observed in specimens sintered at 1650°C , well below the eutectic temperature,

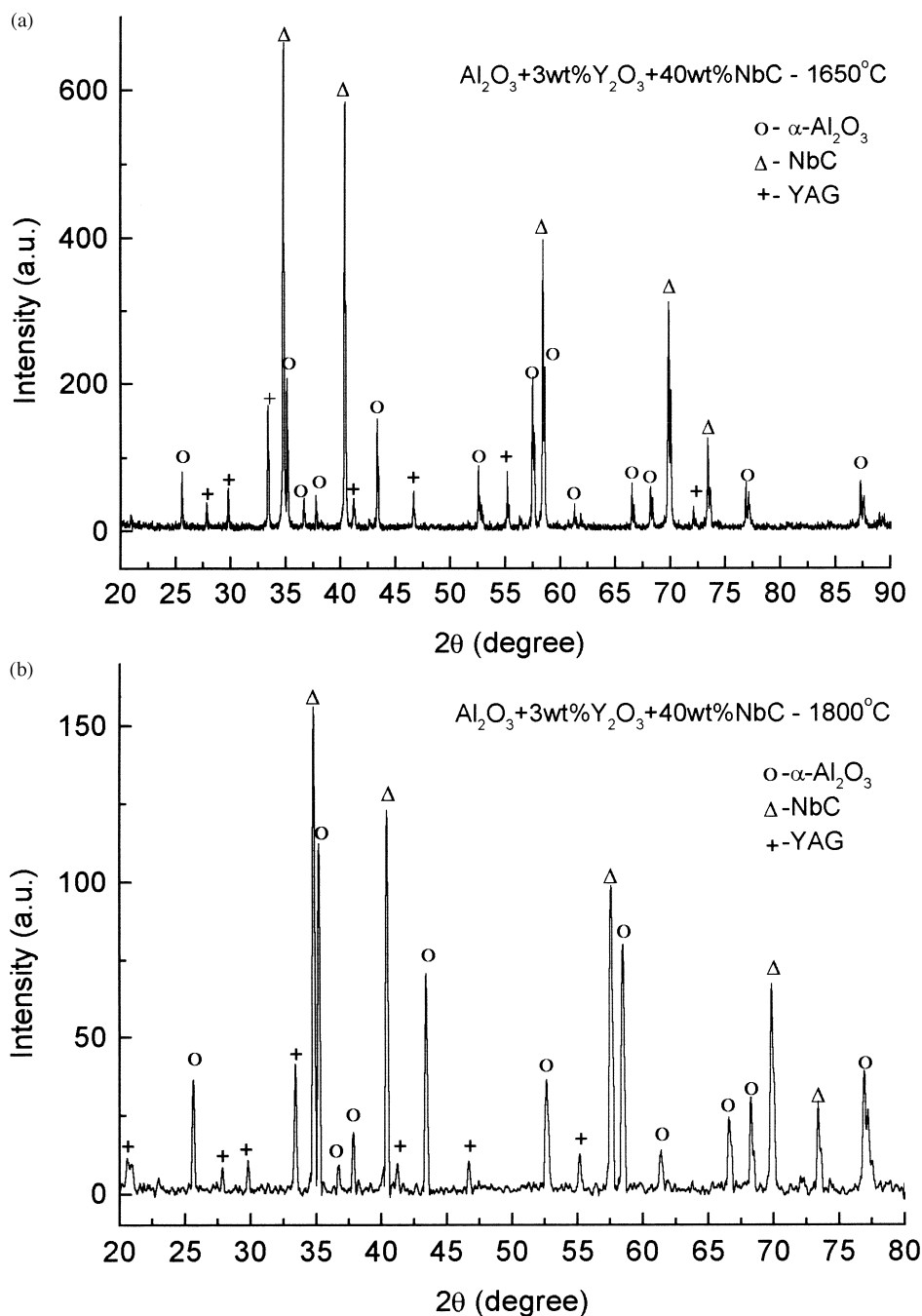


Fig. 6. X-ray diffraction patterns of Al_2O_3 –40 wt.% NbC–3 wt.% Y_2O_3 sintered at (a) $1650^\circ\text{C}/15$ min and (b) $1800^\circ\text{C}/15$ min.

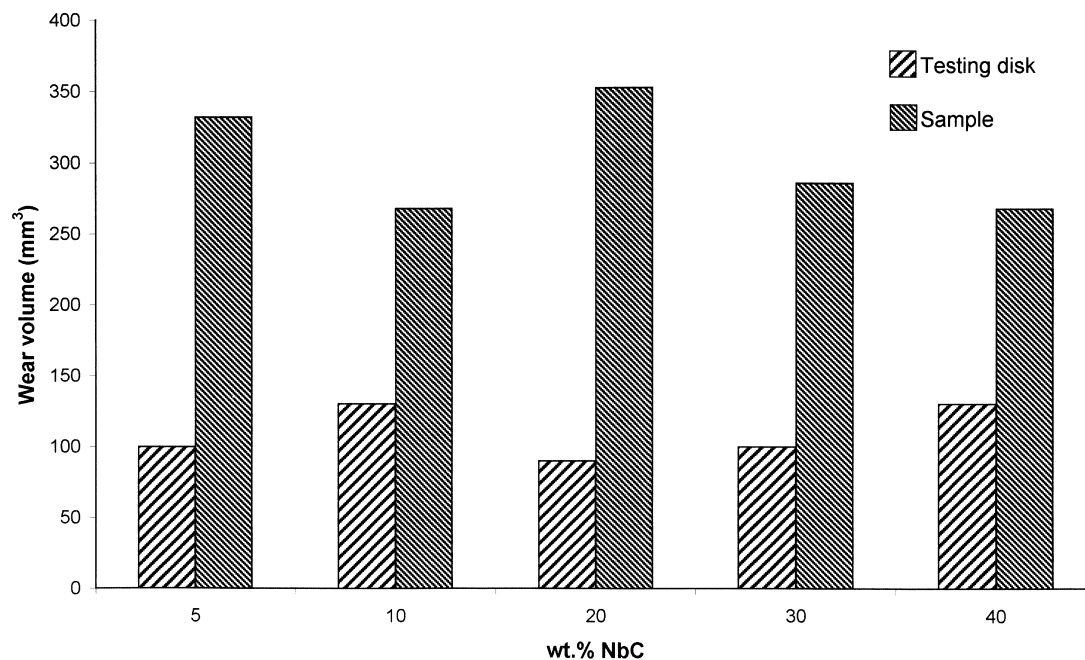


Fig. 7. Wear volume of steel disk and sample from pin-on-disk tests of $\text{Al}_2\text{O}_3\text{--NbC--Y}_2\text{O}_3$ composites as a function of NbC contents.

suggests that YAG can be formed from a solid-state reaction involving Y_2O_3 and Al_2O_3 , and that it does not depend on an eutectic reaction.

Increasing the sintering temperature from 1650 to 1800°C improved the hardness of the composite. Whereas sintering at 1650°C resulted in a maximum hardness of 12 GPa, specimens sintered at 1800°C depicted hardness values in the range 15.5–16.8 GPa. Increasing the NbC contents from 5 to 40 wt.% did not result in any significant improvement in hardness, regardless of the sintering temperature. Apparently, increasing the NbC contents as a means of improving hardness is only effective in highly dense materials (>99% TD), which are particularly challenging to obtain, especially for high contents of refractory carbides dispersed in alumina [4,10]. Within the margin of error, these values are somewhat compatible to those obtained from hot-pressing similar composites [17] to 98% TD, and adequate for metal working [6].

The fracture toughness of $\text{Al}_2\text{O}_3\text{--NbC}$ composites did not change significantly within the margin or error with either sintering temperature or NbC contents. Average values ranged from 2.8 to 4.2 $\text{MPa m}^{1/2}$. Contrary to what was observed for the hardness of the material, the fracture toughness did not vary with density. The present values for fracture toughness are in good agreement with those obtained for similar Al_2O_3 composites reinforced with refractory carbides, such as $\text{Al}_2\text{O}_3\text{--NbC}$ sintered without additives (2.5–4.5 $\text{MPa m}^{1/2}$) [17] and $\text{Al}_2\text{O}_3\text{--TiC}$ (3.4–4.5 $\text{MPa m}^{1/2}$) [5,8]. Although cemented carbides (10–25 $\text{MPa m}^{1/2}$) [5,18,19] and cemented carbide–ceramic composites (6–10 $\text{MPa m}^{1/2}$) [2,19] are tougher, these materials lack intrinsic advantages of

ceramic–ceramic composites with respect to hot hardness, fast cutting speeds, thermal stability and wear resistance.

The wear resistance of $\text{Al}_2\text{O}_3\text{--NbC--Y}_2\text{O}_3$ composites was also evaluated (Fig. 7). Raising the NbC contents from 5 to 40 wt.% did not promote any significant improvement in the wear resistance of the composite, represented by an oscillating behaviour of the wear volume of both disk and sample. Higher wear volumes were observed from the composite samples compared to the disk counterpart, which may be related to the relative low density of the composites (~97% TD). Similarly, no improvement was observed increasing the TiN content from 5 to 25 vol.% in Al_2O_3 -reinforced composites [20], characterised by high relative densities ($\geq 99\%$ TD). Therefore, a suggested composition for future studies would be $\text{Al}_2\text{O}_3\text{--}(20\text{--}40 \text{ wt.}\%) \text{NbC--}3 \text{ wt.}\% \text{Y}_2\text{O}_3$ as it combines reasonably good hardness (~17 GPa) and fracture toughness (~4 $\text{MPa m}^{1/2}$). Further investigations are still required in order to develop denser $\text{Al}_2\text{O}_3\text{--NbC}$ composites characterised by optimised fracture toughness combined with high hardness and wear resistance. Liquid phase sintering with Y_2O_3 additions is capable of producing cost-efficient $\text{Al}_2\text{O}_3\text{--NbC}$ composites capable of withstanding service conditions in metal cutting and machining.

4. Conclusions

The addition of 3 wt.% Y_2O_3 to $\text{Al}_2\text{O}_3\text{--NbC}$ composites promoted the formation of a liquid phase which enhanced the densification of the material sintered at

1800°C, above the Al_2O_3 – Y_2O_3 eutectic temperature (1760°C). Sintering at 1650°C resulted in lower densities which can be attributed to a solid-state sintering mechanism. Materials sintered in the presence of liquid phase showed hardness and toughness around 17 GPa and 4 MPa $\text{m}^{1/2}$, respectively. The wear resistance did not change significantly with NbC contents. Al_2O_3 –3 wt.% Y_2O_3 reinforced with 20–40 wt.% NbC are potential candidates for cutting tool materials.

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