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Abstract

This study investigated effects of cryogenic treatment and post tempering on microstructure parameters and mechanical properties of cemented tungsten carbide for die applications. Carbide grain size was increased after the cryogenic treatment resulted in an increased carbide contiguity. After tempering process, tungsten carbide grain size was reduced and more uniform in size distribution with less in WC-Co debonding. The fracture toughness of cryogenically treated carbide which evaluated from microstructure parameter was slightly increased from 20 to 21.1 $\mathrm{MPa \cdot m^{1/2}}$. After tempering, the fracture toughness was almost unchanged compared to that of untreated sample and the number of tempering cycles did not affect the fracture toughness. However, hardness and scratch resistance were improved 10% and 60%, respectively, by cryogenic treatment. Increasing the number of post tempering cycles after applying the cryogenic treatment result in reducing hardness but did not affected the scratch resistance of the cemented tungsten carbide. In addition, results of ball on disc tribological testing indicated that wear resistance of the cemented tungsten carbide could be improved by cryogenic treatment due to the higher hardness and scratch resistance as expected. The wear volume was increased with the number of tempering cycles. Therefore, sliding wear resistance and scratch resistance applications in which material surface failure behavior is mainly controlled by surface hardness, post tempering process might not be required.

Keywords: cemented tungsten carbide, cryogenic treatment, post tempering, fracture toughness, wear resistance

1. Introduction

Cemented tungsten carbides or hard materials are wildly used in hard and high-quality wear resistance applications. Beside the use for cutting tools, they are also used as moderate-impact and high-impact dies such as wire drawing, bunching, stranding, guiding, and split die applications (Davis, 1995). This is dues to hardness, strength and fracture toughness of the materials. It is well known that the mechanical properties of the cemented tungsten carbide mainly depend on an average grain size, cobalt content and processing to achieve microstructure (Gopal, 1998; Ndlovu, 2009). Typical microstructure of the cemented tungsten carbide composed of WC, TiC, TaC embed in the cobalt binder matrix. In addition, a ternary compound of tungsten, cobalt and carbon (η phase) can exist during the cooling state of sintering (Gopal, 1998). Previous studies indicate that using cryogenic treatment was mainly on improving wear resistance whereas life of the tungsten carbide cutting tools was well documented (Arun, Arunkumar, Vijayaraj and Ramesh, 2018; Gill, Singh, and Singh, 2012; Sreerama, Sornakumar, Venkatarama, Venkatram, 2009; Young, Seah and Rahman, 2006). The cryogenic treatment is generally performed in two methods, shallow and deep practice. The shallow cryogenic treatment is executed at temperatures in the range -80^{o}C whereas deep cryogenic treatment is carried out at temperatures between -140 and -196 ^{o}C (Baldissera and Delprete, 2008; Kalsi, Sehgal and Sharma 2010). The cryogenic treatment process involves slowly cooling materials to the cryogenic, holding it for a certain period of time and then gradually warming back to room temperature. Controlled cryogenic treatment increases values of fineness, uniforms distribution and densification of cobalt binder. It does not affect crystal structure of tungsten carbide (WC) particles and consequently, it improves hardness and bending strength of the cemented carbide (Razavykia, Delprete and Baldissera, 2019). Overall the treatment makes the carbides more firm and better wear resistance. Dhande, Kane, Dhobe and Gogte (2018) studied the effect of soaking periods in cryogenic treatment of tungsten carbide insert. They found that 8 hrs. soaking period at -185 ^{o}C brought about the improvement in the wear characteristics that obtained by pin on disc wear test. Ozbek, Cicek, Gulesin and Ozbek, (2014) showed that the carbide inserts which cryogenically treated at −145 °C for 24 hrs. exhibited the best wear resistance in turning stainless steel. Formation of W_2C and Co_3W_3C (secondary carbides), alongside with cobalt binder create a compressive residual stress and tough matrix (Gill et al., 2012). These can improve wear resistance of the carbide cutting tools. Ozbek, Cicek, Gulesin and Ozbek, (2016) stated that the treated inserts in dry turning stainless steel showed a better performance than the untreated ones of up to 34% and 53% in terms of frank wear and crater wear, respectively. Arun et al., (2018) mentioned that deep and shallow cryogenically treated drill exhibit equivalent performance in drilling small series of holes. As a result, shallow cryogenically treated drill would be more economical working. In addition, effect of tempering followed cryogenic treatment has been studied together with the tungsten carbide inserts (Deshpane and Venugopal, 2018; Kalsi Sehgal and Sharma, 2010; Thakur, Ramamoorthy and Vijayaraghavan, 2008). Deshpande et al. (2018) and Kalsi et al. (2014) found that tempering had a significant influence on the phases presented in WC-Co inserts without changing the surface morphology and subsequently influenced their machining performance. The cryogenic treatment improved the microhardness of the tungsten carbide inserts but the microhardness was decreased with an increase in the number of tempering cycles after the treatment (Kalsi et al. 2014). In addition, the cryogenically treated insert with double and triple tempering cycles exhibited the highest wear resistance. However, Kalsi et al. (2014) indicated that the fourth tempered insert reduced effect of cryogenic treatment by disruption in the cobalt binder. Recently, Weng, Gu, Wang, Liu, Cai and Wang (2019) found that deep cryogenic treatment could improve both fracture toughness and wear resistance of WC-20Co cemented carbide for die applications. However, scientific literature in the area of improvement mechanical properties of the cemented tungsten carbide for die applications is limited. No study is evident about effectiveness of post tempering and number of tempering cycles on microstructure and mechanical properties of the cemented tungsten carbide in case of die applications that emphasize fracture toughness and high wear resistance. The objectives of this investigation are to evaluate the effects of cryogenic treatment and post tempering cycles on microstructure parameter and mechanical properties; hardness, scratch resistance and wear analysis of the cemented tungsten carbide for die applications.

2. Materials and Methods

2.1 Materials

The cemented tungsten carbide (80 wt.%WC- 20 wt.%Co) were used as samples of this study and classified as highly impact tool and die applications.

2.2 Methods

The samples were cryogenically treated with step cooling at -140 ℃ and held at this temperature for 24 hrs. After the cryogenic treatment (CT), the samples were gradually brought to room temperature, then submitted to different tempering treatments; single, double and triple tempering at 200 ℃ for 2 hrs. to relieve stresses induced in cryogenic treatment.

2.3 Microstructure Analysis

Microstructure analysis of the WC-Co cemented carbide was carried out under Scanning Electron Microscope (JEOL, Japan). The samples were polished by diamond paste (1 μm) to reach mirror finish and etched with KOH reagent. To study effects of the cryogenic treatment, microstructure parameters were evaluated. This included carbide grain size distribution, average carbide size, carbide contiguity (WC/WC interface), and binder mean free path for each sample. All of these were evaluated from the 9 SEM photomicrographs. Fracture toughness, K_{1C} of the WC-Co cemented carbide was calculated by these microstructure parameters.

2.4 Hardness Test

Measurements of microhardness was performed under a load of 300 g for 15 s by using a micro-vickers hardness tester (FUGITEC, Japan). The microhardness was measured at 13 points for each sample and mean value of these measurements was accepted as microhardness value of the samples.

2.5 Scratch Test

Scratch resistance of the samples was carried out on a scratch tester (CSM Instruments, Switzerland) with diamond indenter and with applied progressive loads from 0.9 N to 100 N. Scratching speed was 10 mm/min and scatch length was 10 mm.

2.6 Wear Test

Sliding wear test was performed using a Ball on disc tribological testing machine (CSM Instruments, Switzerland) at room temperature under non-lubricated condition. The cemented tungsten carbides were prepared as the disc and the ball made of cemented tungsten carbide coated with AlCrTiN. A load of 15 N was imposed that created the maximum 3,050 MPa in contact pressure. Sliding velocity was fixed at 0.1 cm/s, while cycles were changed from 1500 to 2000, 2500, 3000, and 3500. A contact radius of 3 mm was applied. A profilometer was used to map the contour of the worn surfaces. Wear volume and rate of wear were subsequently determined.

3. Results and Discussion

3.1 Microstructural analysis and fracture toughness evaluation

Figure 1 shows SEM pictures of the untreated, the cryogenically treated and the cryogenically treated and single tempered cemented tungsten carbide samples. Carbide grain size is one factor that affects the mechanical properties and wear resistance. To study the effect of cryogenic treatment, sizes of carbide grains were randomly measured by using the line intercepts method from 9 images, 15 grains per image, in order to compare the average grain size of the untreated with cryogenically treated samples. The average of carbide grain sizes of the samples and standard deviation with various conditions were shown in Figure 2. Upon measuring sizes of the carbide grains, it was found that the average grain size of the untreated cemented tungsten carbide was 4.32 μm and the cryogenically treated cemented tungsten carbide was 4.5 μm, respectively. Cryogenic treatment led to an increase of 4% in the grain size and greater standard deviation of the treated cemented tungsten carbide with respect to that of the untreated one. As mention by Thornton (2014), the increasing of tungsten carbide grains reduces micro porosity and results in a more contiguous microstructure. The larger grain size caused an increase in carbide contiguity. This agree well with that found in case of the tungsten carbide insert tool (Nursel, 2016). However, the grain size was slightly reduced to 4.30 μm after the tempering and the standard deviation was also reduced. This can be interpreted that the carbide of the sample is more uniform in size after the tempering. Increasing in the number of tempering cycles does not affect the carbide grain size but it becomes more uniform in size distribution. Microstructure of the untreated and the cryogenically treated cemented tungsten carbide was shown in Figure 1(a) and 1(b), respectively. It can be observed that WC debonding and fragment of WC grains became less in cryogenically treated sample. In case of the single tempered sample as illustrated in Figure 1(c), the WC debonding is hardly found but small torn off WC are existed. Microstructure parameters which are the carbide contiguity and binder mean free path were evaluated and also shown in Table 1. The carbide contiguity indicated the degree of contacts between carbide grains. It was found that cryogenic treatment increased the carbide contiguity from 0.82 to 0.87. These might be a result of the cobalt binder contraction related to the cryogenic treatment. Consequently, the carbide particles become closer. In case of the single tempered sample, carbide contiguity was unchanged, comparing with the cryogenically treated one. The cobalt binder was slightly expanded during the double and triple tempering, and the carbide contiguity was reduced to 0.81.

Fracture toughness, K_{1C} of the cemented tungsten carbide, is known to increase as binder phase volume fraction and average carbide grain size. The binder phase mean free path is increased, while decreased carbide contiguity causes a decrease in the fracture toughness. K_{1C} of the cemented carbide can be evaluated from microstructure parameters, as suggested by Gopal (1998); Mari, Llanes and Nebel (2014):

$$K_{IC} = 3.907 + 0.325V_B\% + 2.389\bar{d}_C - 0.878\lambda + 2.065C$$

$V_B\%$, $\bar{d}_C$, λ and C are vol% binder phase, average carbide grain size, binder phase mean free path and carbide contiguity, respectively. The fracture toughness, K_{1C} of the samples as evaluated from the microstructural parameters are also shown in Table 1. The K_{1C} of the cryogenically treated tungsten carbide was higher than that of the untreated tungsten carbide. These might be due to the increased grain size and carbide contiguity which are the microstructure parameter which promoted the K_{1C}. After the single tempering, the fracture toughness was almost unchanged and the number of tempering cycles did not affect the fracture toughness but the microstructure became more uniform.

3.2 Hardness

Hardness of the untreated and cryogenically treated cemented tungsten carbide are shown in Table 2. The data indicates that, average hardness of the untreated sample is 989.02 HV and the cryogenically treated sample is 1079.93 HV with high value of standard deviation. Since, the tempering process affected hardness of the cryogenically treated samples, hardness of the cryogenically treated sample was significantly reduced after the single tempering. Standard deviation of the hardness was also reduced after tempering process. This indicated that: the tempered samples are more uniform in hardness; increasing the number of tempering cycles bring about the slightly decreasing in hardness; and tempering process reduces residual stress without changing the microstructure.

3.3 Scratch Test

Studying effects of the cryogenic treatment, fracture toughness and hardness on the scratch resistance of the cemented tungsten carbide were evaluated in terms of critical loads for the appearance of first cracks (L_{C1}), dense cracking pattern (L_{C2}) and completed cracks (L_{C3}) (Paiva et al., 2018). The critical loads under the scratch test are shown in Figure 3. The scratch resistance properties of the cemented tungsten carbide could be improved by the cryogenic treatment and the tempering process. First crack in the cryogenically treated samples could not observed up to the critical load (L_{C1}) of 56.02 N and dense cracking pattern could not be detected up to the maximum load of 66.96 N. The fracture crack occurred at 88.81 N. These evidence higher scratch resistance of the treated tungsten carbide comparing with the untreated cemented tungsten carbide that the first crack was occurred at the critical load (L_{C1}) of 35.42 N. This is because of the reduced WC debonding and fragment of WC grains in cryogenically treated sample as illustrated in Figure 1(b). In case of the cryogenically treated and single tempered tungsten carbide which less WC debonding and fragment, earlier cracks were observed up to the critical load (L_{C1}) about 55 N which closed to the L_{C1} of the cryogenically treated samples. The number of tempering cycles did not cause much changes in the critical loads for the appearance of the first crack. However, the cryogenically treated samples were completely cracked under the highest load. These might be due to the highest hardness of the cryogenically treated sample. The experimental results were consistent with WC-Co carbide with a high cobalt content. The dominant damage mechanisms are the plastic deformation of the WC grains via slip and the formation of intergranular cracks

(Duszova A.N., Casnadi T., Sedlak R., Hvizdos P. and Dusza J., 2019). Therefore, the scratch resistance of the cemented tungsten carbide could be improved by the cryogenic treatment and tempering cause that the debonding of tungsten carbide decreased.

3.4 Wear resistance

An illustration of the effect of the cryogenic treatment and tempering on wear volume of the cemented tungsten carbide is shown in Figure 4. The wear volume was increased as the number of the test cycles. The lowest wear volume was obtained in the case of cryogenically treated cemented tungsten carbide with the high hardness of 1074.93 HV and also displayed the highest fracture toughness 21.1 $\text{MPa} \cdot \text{m}^{1/2}$. This is due to the densification of the cobalt binder, which induces a compressive stress on the WC grains, resulting in an increase in wear resistance. The wear volume of the cryogenically treated with single tempered tungsten carbide was slightly greater than the cryogenically treated one. However, increasing the number of tempering cycles brought about the increasing wear volume. The cryogenically treated with triple tempered cemented tungsten carbide showed the highest wear volume which closed to the wear volume of the untreated cemented tungsten carbide. These might be a result of the triple tempering after the cryogenic treatment produced disruptions in the cobalt binder as found in case of carbide insert tool (Kalsi et al, (2014). An analysis of wear scars, the main wear mechanism or abrasive wear is shown in Figure 5. Increasing fracture toughness on the expense of hardness did not affect wear mechanism. Wear rate or volume loss per unit distance was also evaluated as shown in Figure 6 which illustrates the same trend as the wear volume. The cryogenically treated cemented tungsten carbide has the lowest wear rate as compared with the others samples.

4.Conclusions

The study investigated effects of cryogenic treatment and post tempering cycles on microstructure parameters and mechanical properties of cemented tungsten carbide for die applications. It was found that cryogenic treatment and single tempering process promoted a densification of cobalt binder. Single tempering after cryogenic treatment promoted the uniform grain size and carbide contiguity of the cemented tungsten carbide. The hardness and fracture toughness, K_{1C} of the cryogenically treated tungsten carbide, were increased with increasing mean free path in the cobalt binder phase and increasing carbide contiguity. Hardness of the cemented tungsten carbide decreased with an increase in the number of tempering cycles after applying the cryogenic treatment. The cryogenically treated tungsten carbide exhibited the highest scratch resistance due to higher surface hardness. Cryogenic treatment improved wear resistance of the cemented tungsten carbide, however, wear volume increased with the number of tempering cycles. The cryogenically treated with triple tempered cemented tungsten carbide showed the highest wear volume which closed to the wear volume of the untreated cemented tungsten carbide. The investigation indicated that hardness is still the main factor affecting sliding wear resistance under the current testing conditions and surface scratch resistance. Tempering after the cryogenic treatment does not increase fracture toughness but result in lower hardness. However, post tempering could promote a uniform in microstructure and hardness could be advantaged for die application with impact wear and high local stress area. For sliding wear resistance and scratch resistance applications in which material surface failure behavior is mainly controlled by surface hardness, post tempering process might not be required.

References

Arun M., Arunkumar N., Vijayaraj R. and Ramesh B. (2018). Investigation on the performance of deep and shallow cryogenic treated tungsten carbide drills in austenitic stainless steel. *Measurement*, (125), 687-693. doi:10.1016/j.measurement.2018.05.018

Baldissera, P. and Delprete C. (2008). Deep cryogenic treatment: A bibliographic review, *The Open Mechanical Engineering Journal*, (2), 1-11. doi:10.2174/1874155X00802010001

Davis, J. R. (1995). *ASM Specialty Handbook: Tool Materials*, Ohio U.S.A, *ASM International.*

Deshpande R.G. and Venugopal K.A. (2018). Machining with cryogenically treated carbide cutting tool inserts. Materialstoday: proceedings, 5(1), 1872-1878. doi:10.1016/j.matpr.2017.11.288

Dhande S.T., Kane V.A., Dhobe M.M., and Gogte C.L. (2018). Influence of soaking periods in cryogenic treatment of tungsten carbide. *Procedia Manufacturing*, (20), 318-328. doi:10.1016/j.promfg.2018.02.047

Duszova A.N., Casnadi T., Sedlak R., Hvizdos P. and Dusza J. (2019). Small-scale mechanical testing of cemented carbides from the micro-to the nano-level: A review. *Metals*, 9(502). doi:10.3390/met9050502

Gill S.S., Singh J., Singh H. and R. Singh (2012). Metallurgical and mechanical characteristics of cryogenically treated tungsten carbide (WC–Co). *International Journal of Advanced Manufacturing Technology*, (58), 119-131. doi:10.1007/s00170-011-3369-4

Gopal S. U. (1998). Microstructural aspects of cemented carbide, *Cemented tungsten* *carbides: production, properties and testing* (pp.166-192). New Jersey, U.S.A. Noyes Publication.

Kalsi N.S., Sehgal R. and Sharma V. S. (2010). Cryogenic treatment of tool materials: A review. *Materials and Manufacturing Processes*, (25), 1077-1100. doi:10.1080/10426911003720862

Kalsi N.S., Sehgal R. and Sharma V. S. (2014). Effect of tempering after cryogenic treatment of tungsten carbide-cobalt bounded inserts. *Bulletin of Materials Science*, 37(2), 327-335. doi:10.1080/10426911003720862

Mari D., LLanes L and Nebel E. (2014). Fracture and strength of hardmetals at room temperature, *Comprehensive Hard materials*, (pp.301-344). Elsevier

Ndlovu S. (2009). *The wear properties of tungsten carbide-cobalt hardmetals from the nanoscale up to the macroscopic scale* (Doctoral thesis, University of Erlangen-Nuremberg, Erlangen, Germany). Retrieved from https://www.researchgate.net/publication/296062262_The_Wear_Properties_of_Tungsten_Carbide_Cobalt_Hardmetals_from_the_Nanoscale_up_to_the_Macroscopic_Scale

Ozbek, N., Cicek, A., Gulesin, M. and Ozbek, O. (2014) Investigation of the effects of cryogenic treatment applied at different holding times to cemented carbide inserts on tool wear. *International Journal of Machine Tools & Manufacture*, (12), 34-43. doi:10.1016/j.ijmachtools.2014.06.007

Ozbek, N., Cicek, A., Gulesin, M. and Ozbek, O. (2016). Effect of cutting conditions on wear performance of cryogenically treated tungsten carbide inserts in dry turning of stainless steel. *Tribology International*, (94), 223-233. doi:10.1016/j.triboint.2015.08.024

Paiva J. M., Fox-Rabinovich G., Junior E. L., Stolf P., Ahmed Y. S., Martins M. M., Bork C. and Veldhuis S. (2018). Tribological and wear performance of nanocomposite PVD hard coatings deposited on aluminum die casting tool, Materials, 11(358), 1-15. doi:10.3390/ma11030358

Razavykia A., Delprete C. and Baldissera. (2019). Correlation between microstructural alteration, mechanical properties and manufacturability after cryogenic treatment: A review, *Materials*, 12(3302), 1-36. doi:10.3390/ma12203302

Sreerama R., Sornakumar T., Venkatarama R. and Venkatram M. (2009). Machinability of C45 steel with deep cryogenic treated tungsten carbide cutting tool inserts. *International Journal of Refractory Metals & Hard Materials*, (29), 181-185. doi:10.1016/j.ijrmhm.2008.04.007

Thakur D., Ramamoorthy B. and Vijayaraghavan L. (2008), Influence of different post treatments on tungsten carbide–cobalt inserts. *Materials Letters,* **(**62), 4403-4406. doi:10.1016/j.matlet.2008.07.043

Thornton R.W. (2014). *Investigating the effects of cryogenic processing on the wear performance and microstructure of engineering materials (Doctoral thesis,* The University of Sheffield, U.K.). Retrieved from https://pdfs.semanticscholar.org/234f/4046cca6d2e0850ec75dc90d9aa34281ae97.pdf

Weng Z., Gu K., Liu X., Cai H. and Wang J. (2019) Effect of deep cryogenic treatment on the fracture toughness and wear resistance of WC-Co cemented carbides. International Journal of Refractory Metals and Hard Materials, (85), doi:10.1016/j.jirmhm.2019.105059

Yong A.Y.L., Seah K.H.W., and Rahman M. (2006) Performance evaluation of cryogenically treated tungsten carbide tools in turning. *International Journal of Machine Tools & Manufacture*, (46), 2051-2056. doi:10.1016/j.ijmachtools.2006.01.002

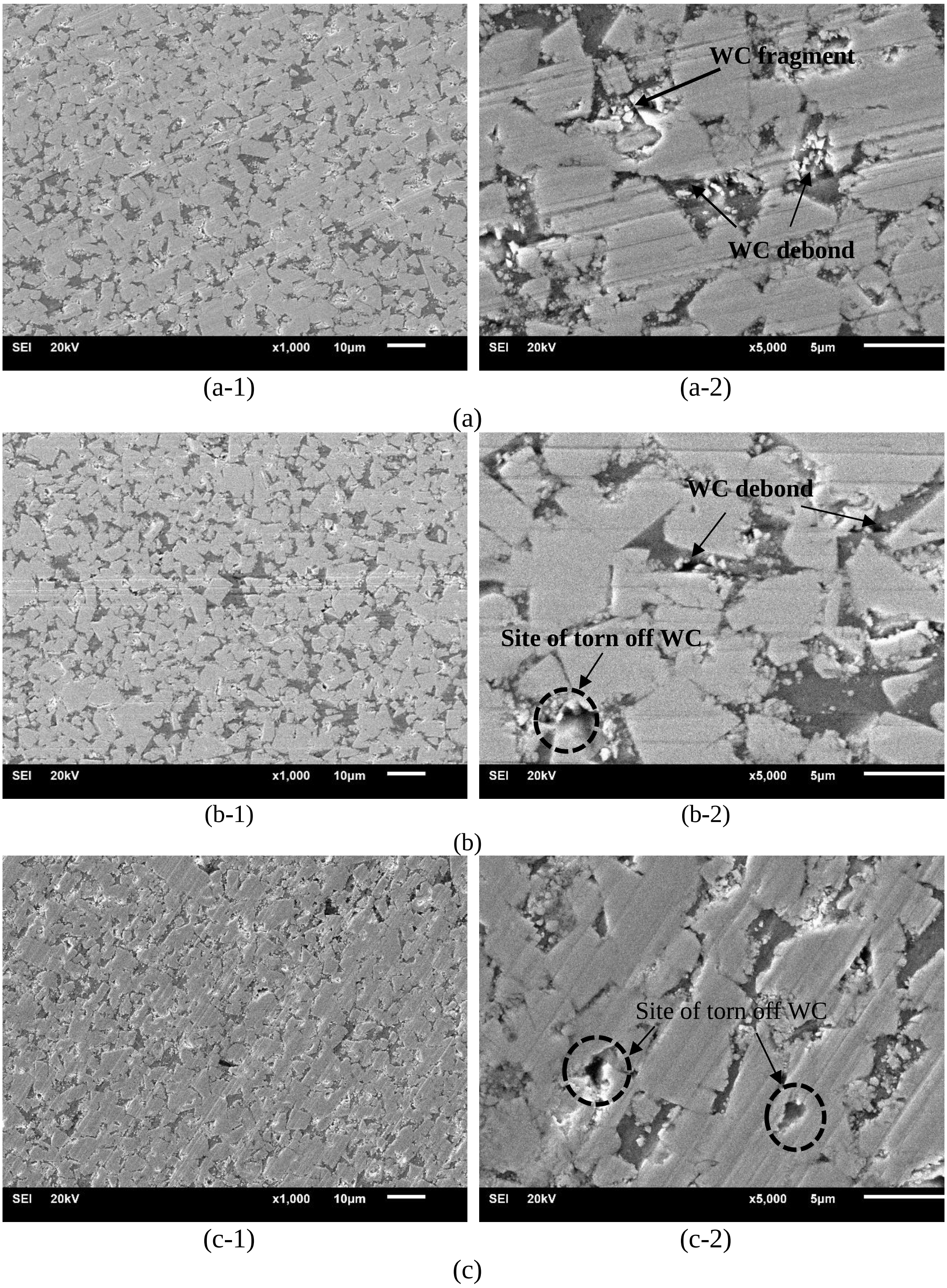


Figure 1. Microstructure of cemented tungsten carbide (a) untreated (b) cryogenically treated (c) cryogenically treated with single tempered.

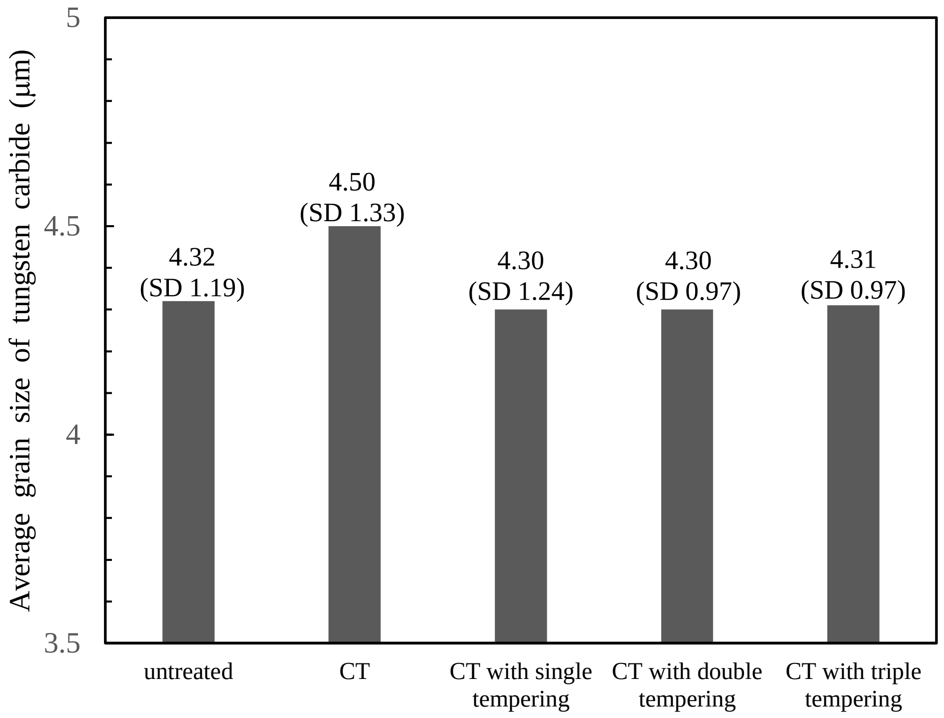


Figure 2. Effect of cryogenic treatment and tempering on average grain size of tungsten carbide.

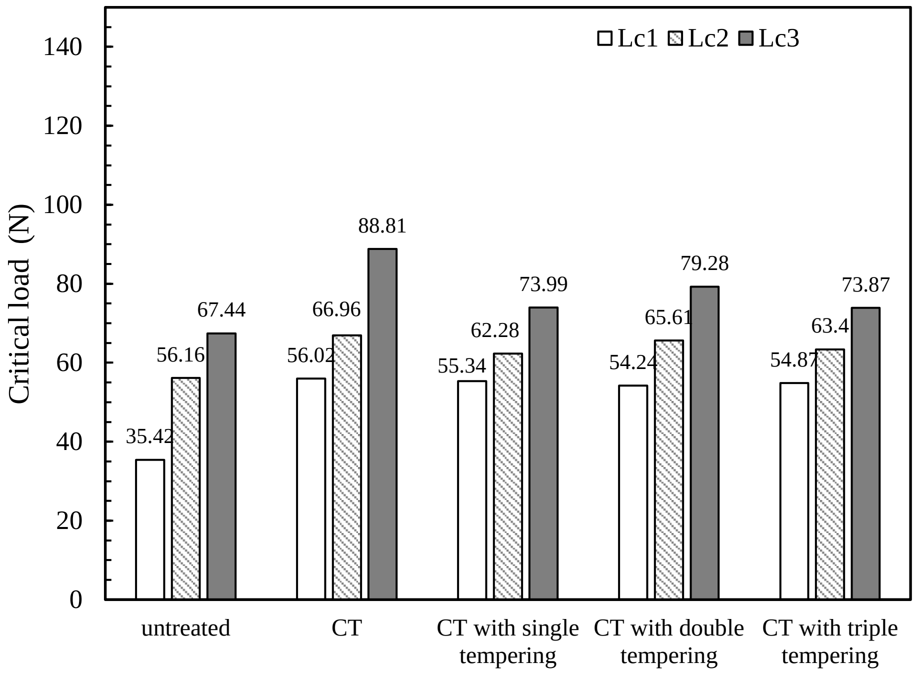


Figure 3. Effect of the cryogenic treatment and tempering on critical load under scratch resistance test of cemented tungsten carbide.

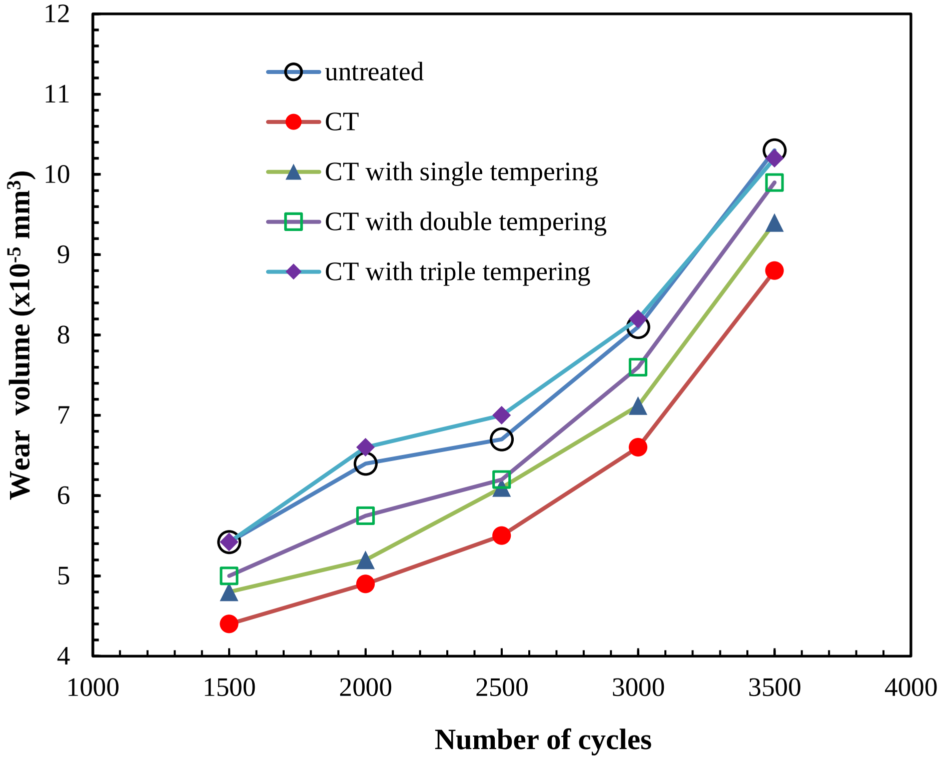
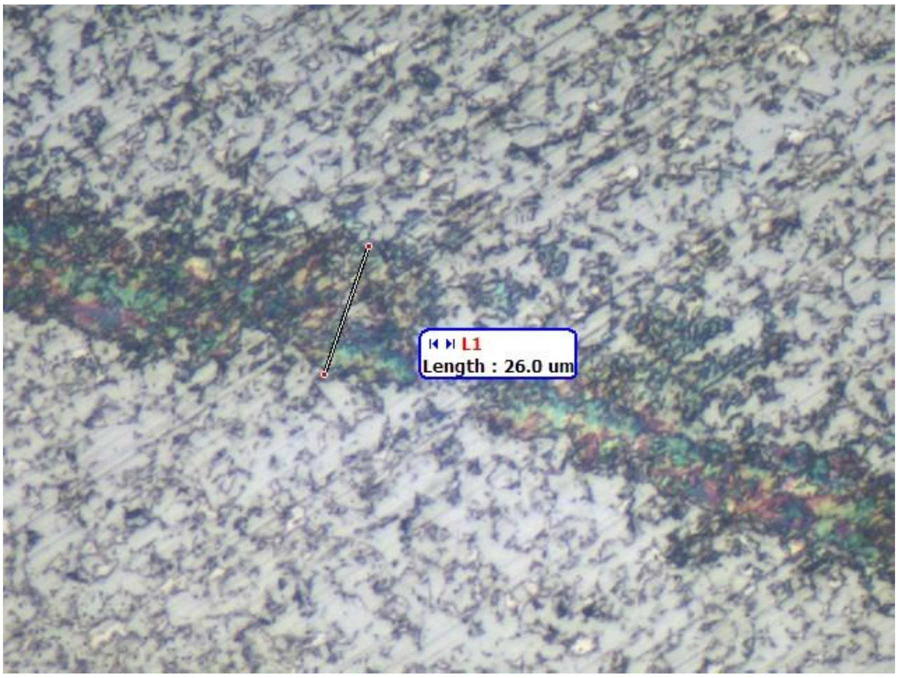
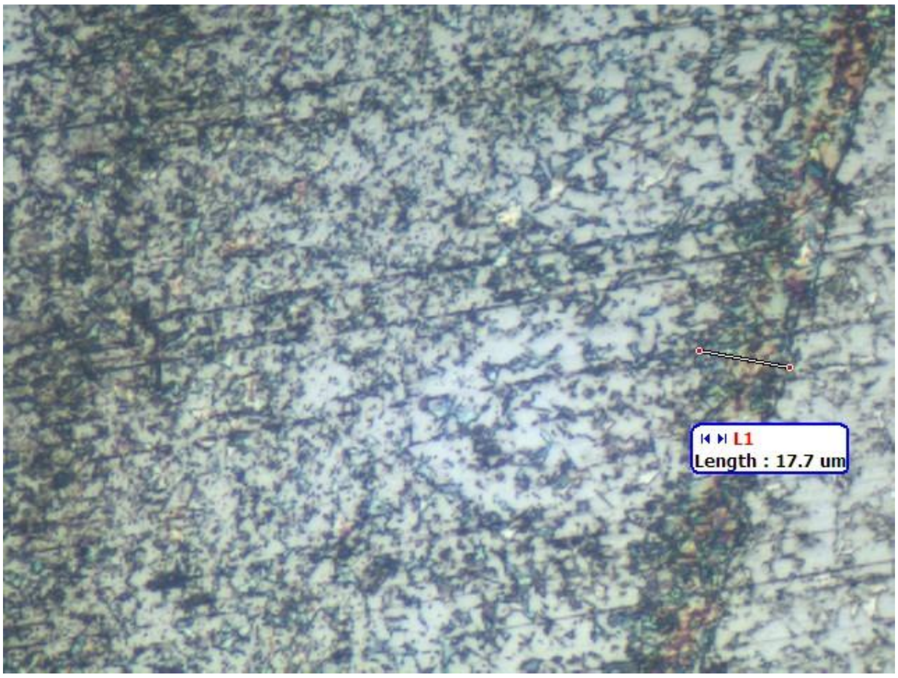


Figure 4. Evolution of wear volume with number of cycles of cemented tungsten carbide.



(a)



(b)

Figure 5. Wear scar after ball on disc tribological test for (a) untreated (b) cryogenically treated tungsten carbide.

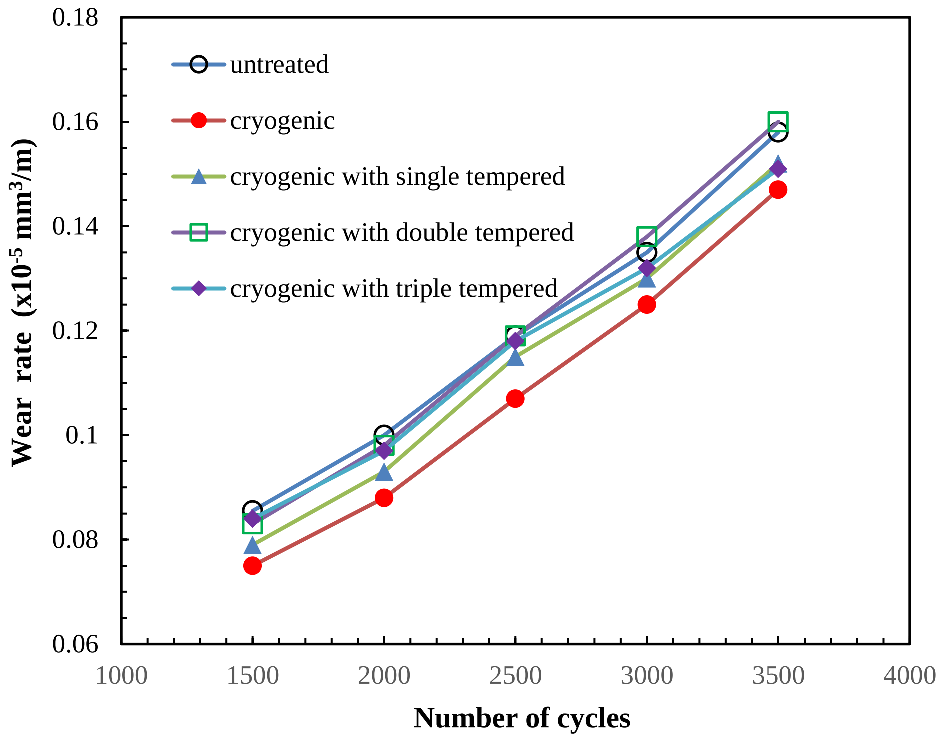


Figure 6. Wear rate versus number of cycles of cemented tungsten carbide.

Table 1 Microstructural parameter and K_{1C} of cemented tungsten carbide.

Condition	Carbide average size (µm)	Carbide contiguity	Binder mean free path (µm)	K_{1C} ($MPa \cdot m^{1/2}$)
Untreated	4.32 ± 1.19	0.82	1.97	20.7
Cryogenic treated	4.50 ± 1.33	0.87	2.11	21.1
Cryogenic treated and single tempered	4.30 ± 1.24	0.87	2.01	20.7
Cryogenic treated and double tempered	4.30 ± 0.97	0.81	1.94	20.6
Cryogenic treated and triple tempered	4.31 ± 0.97	0.81	1.95	20.7

Table 2 Hardness of the cryogenically treated and tempered cemented tungsten carbide.

Condition of tungsten carbide	**Hardness (HV)**	**Standard deviation**
Untreated	989.02	23.04
Cryogenically treated	1074.93	32.66
Cryogenically treated with single tempered	962.47	14.61
Cryogenically treated with double tempered	951.81	15.51
Cryogenically treated with triple tempered	947.86	7.89