

The Effect of Deep Cryogenic Treatment on Microstructure, Hardness and Wear Behavior of INDRA 5 Cold Work Tool Steel

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Abstract

In this study, the effect of cryogenic treatment on microstructure, hardness and wear behavior of cold work tool steel containing 5 wt. % Cr used in the production of cold rollers was investigated. For this purpose, the starting and finishing temperatures of the martensite transformation were determined by dilatometer test (234 °C and -102 °C, respectively). Then, quenched-tempered heat treatment (QT) and quenched-cryogenic heat treatment for 24 hours-temper (DCT) were performed on the samples. In the next phase, microstructure of the samples was evaluated by X-ray diffraction (XRD) and scanning electron microscopy (SEM), and their hardness and wear behavior were studied by pin-on-disk wearing machine. In order to determine the wear mechanisms, the level of the wear effect was investigated by SEM. The obtained results showed that in deep cryogenic treatment compared to the quenched-tempered treatment, due to the transformation of retained austenite to martensite, precipitation of fine carbides, increased volume fraction of carbides and more appropriate distribution of these carbides, led to an increase in hardness and wear resistance to 3HRC and 30%, respectively. The study of the worn-out surface and its products showed that the wear mechanism was adhesive wear along with tribochemical wear. Deep cryogenic treatment reduced the amount of adhesive wear.

Keywords: Cryogenic heat treatment; Cold work tool steel; Retained austenite; Wear resistance.

1. Introduction

In the high alloy steels, the retained austenite is a consequence of low martensite finishing temperature (M_f). In these steels, the essential shear stress which is necessary for martensite transformation increased as a result of a high amount of alloying elements, and this leads

to the decrease of martensite transformation temperature. Subsequently, the M_f temperature decreases to below the room temperature in the high carbon or alloyed steels due to this higher amount of required cooling. In other words, the retained austenite is not annihilated by cooling the samples down to the room temperature during quenching ^{1,2}. The retained austenite, γ_{Fe} , is soft and decreases desirable properties like wear resistance and hardness ³. In fact one of the major challenges in the tool steels heat treatment is the minimization or elimination of the amount of γ_{Fe} . Cryogenic treatment as a supplementary heat treatment is performed on some tool steels before tempering for decreasing the residual stress, retained austenite and increasing the wear life ⁴. There are two types of cryogenic treatments generally used: (1) shallow cryogenic treatment between -60 °C and -90 °C; (2)

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deep cryogenic treatment at temperatures below -125°C ⁵). In the latter, after the tempering treatment, secondary and highly fine carbides are formed in the structure in addition to retained austenite to martensite transformation ^{6, 7}). Huang showed that in high-speed steel M2, the carbides volume fraction becomes two-fold due to the effect of deep cryogenic treatment ⁸). Meng reported 110-600% improvement in wear resistance at different speeds by performing cryogenic treatment on Fe-12Cr-Mo-V-1.4C steel using the sample on the wheel test ⁹). The conducted researches show that cryogenic treatment improves hardness ^{10, 11}) and wear resistance ^{6, 7}) and increases the bending strength ¹²), dimensional stability ¹³), fatigue resistance ¹⁴), and fracture toughness ¹²) in the steel pieces. Cold work tool steels are the most famous group of steels for performing this treatment. They are commonly used in die and tool productions, where the highest hardness and wear resistance are the major factors in choosing the heat treatment. This process eliminates or reduces the retained austenite and as a result of that increases the hardness of tools ^{15, 17}). The INDRA 5 steel is a cold work tool steel which was first produced by Isfahan Alloy Steel Complex for manufacturing cold rolling rollers. It is worth mentioning there has not been any research conducted on the cryogenic treatment of this steel. Therefore, in this study, the effect of deep cryogenic treatment on the microstructure, hardness and wear behavior of this steel has been investigated.

2. Research Method

In Table 1, the chemical composition of INDRA5 steel is presented. Chemical composition of the steel was analyzed by an optical emission spectrometer. In order to determine the starting and finishing temperatures of the martensite transformation of this steel, dilatometric tests were conducted using a BHR805A/D dilatometer device. The dilatometer samples were prepared according to the SEP1681 standard in cylinder shapes of 4 mm diameter and 10 mm length. In conventional heat treatment (QT), after a preheat treatment at 620°C for 20 min, the samples were austenitized at 980°C for 20 min under the argon gas atmosphere, and then were quenched in oil at 60°C . In order to investigate the effect of deep cryogenic treatment (DCT), liquid nitrogen at -196°C was used. The samples were cooled down to -196°C immediately after quenching and kept at this temperature for 24 hrs. In order to prevent the thermal steep slope and fine crack formation, the heating and cooling speed of the samples in cryogenic treatment were selected to be $1^{\circ}\text{C}/\text{min}$. A schematic of the deep cryogenic heat treatment cycle performed on the samples is shown in Fig. 1. Some samples were tempered immediately after quenching (QT), and some others were tempered after quenching and cryogenic treatment (DCT). The tempered treatment was performed at 150°C and under the atmosphere of argon for 3 hrs. . Hardness test was performed using Rockwell C method according to the DIN 50103 standard using an EMCO 4U750 machine (Germany). For each sample, hardness measurement was done at three points and the average of obtained hardness values was reported.

Table 1. Chemical composition of INDRA5 steel, (in wt. %).

C %	Si %	Mn%	Cr%	Mo%	P %	S%	Fe%
0.79	0.85	0.25	5.1	0.5	0.005	0.008	Bal.

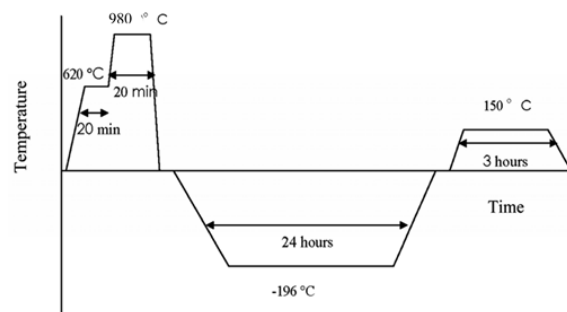


Fig. 1. Schematic presentation of the heat treatment cycle applied on the DCT sample.

Scanning electron microscopy (SEM, Seron AIS-2100) was used to investigate the worn-out surface and microstructure. In order to investigate the changes in the samples microstructure, the samples surfaces were etched using (a) 100 ml of H₂O, 10 g of K₃Fe(CN)₆ and 10 g of NaOH and (b) mixed acid (3 ml of HF, 53 ml of H₂NO₃, 2 ml of CH₃COOH, and 42 ml of H₂O). The etching of the samples in each etchant lasted 2 s and was repeated 3 to 4 times. This results only in the clarification of carbides and does not affect other phases. Afterwards, the surface of the samples surface was analyzed by the use of optical microscopy. To calculate the carbide percentage, the micrographs were analyzed by Clemex Vision (version 3.5.025) image analyzing software. X-ray diffractometer (XRD, Philips PW1730) with Cu-K α radiation was used for determining the phases as well as the retained austenite percentage, which was calculated as per ASTM E975-00 standard. SEM micrographs were used for determination of the carbide percentage. The austenite percentage was calculated as follows:

$$V_{\gamma} = \left[\frac{(1-V_c)(I_{\gamma}/R_{\gamma})}{(I_{\alpha}/R_{\alpha})} + (I_{\gamma}/R_{\gamma}) \right] \quad \text{Eq. (1)}$$

, where V γ and V c are the retained austenite and carbide percentage, respectively, I γ and I α are the integrated intensity per angular diffraction peak (hkl) in the austenite and martensite phases, respectively, and R γ and R α are the austenite and martensite constants, respectively. We can determine the value of R with respect to the (hkl) plane in combination with the phases polarization, multiplicity, and structure factor according to the ASTM E975-00 standard.

In order to investigate the effect of cryogenic treatment on wear resistance, the wear test was performed using pin- on- disk method with a pin of ball bearing steel (AISI 52100) and hardness of 64 HRC and mobile disk (sample) under 120 N load at 0.2 m/s in accordance with the ASTM G99 standard. The wear test was conducted at a sliding distance of 1000 m at a relative humidity of 30 \pm 5% and a temperature of 25 \pm 5 $^{\circ}$ C. The following equation was used to calculate the wear rate:

$$W_r = \Delta m / (\rho \times l \times F) \times 10^6 \quad \text{Eq. (2)}$$

, where W r is the wear rate in mm³ /N.m, Δm is weight loss in kg, ρ is the steel density in gr/cm³, l is the traversed distance in m, F is the applied (loaded) force in N, and the coefficient 106 for unit conversion.

3. Results and Discussion

3. 1. Microstructure

In the dilatometry tests, in order to study phase transformations, the specimens temperature variations and length were simultaneously printed during cooling and heating. According to Fig. 2, after heating, the

samples length increased because of thermal expansion. However, when an α^+ cementite to $\alpha+\gamma$ phase transformation occurred, the specimen length decreased. The starting and finishing temperatures of this transition were 815.2 $^{\circ}$ C (AC₁) and 893.7 $^{\circ}$ C (AC₃), respectively. The specimen length increased again by increasing the temperature. Keeping the sample at 980 $^{\circ}$ C did not result in the change of its length. The length started to decrease as a result of thermal contraction during the quenching process. Upon starting the martensitic transformation at 234 $^{\circ}$ C, the length suddenly increased at a temperature that is called M s . Martensitic transformation stopped when the temperature reached the ambient temperature. The sample was cooled in liquid nitrogen for 15 min at -150 $^{\circ}$ C after being kept at the ambient temperature for 3 min. By decreasing the temperature from ambient, the sample's length decreased and, subsequently, the martensitic transformation at -21 $^{\circ}$ C started again by providing the required driving force with increasing the sample length and continued to -102 $^{\circ}$ C. Then the sample length decreased upon decreasing the temperature. Therefore, the finishing temperature of martensitic transformation (M f) of this steel was determined at -102 $^{\circ}$ C which ended with an increase in the sample length during the cooling process.

Fig. 2b depicts the martensitic transformation region

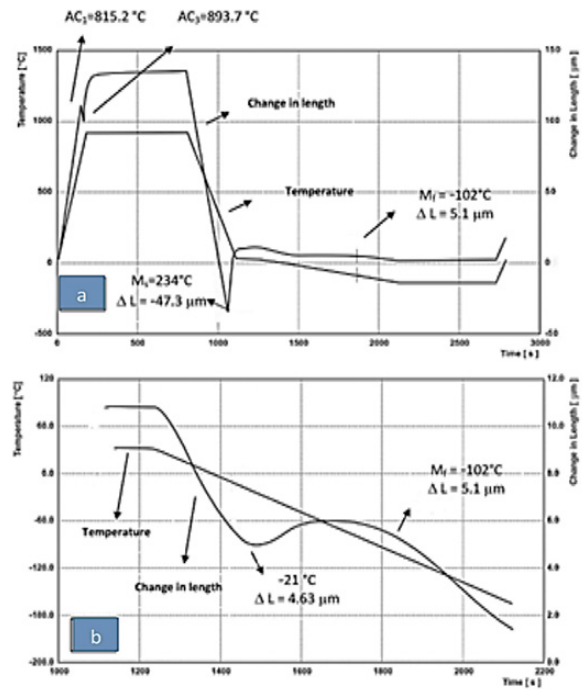


Fig. 2. Dilatometer curve obtained from INDRA5 steel, a) the sample heat treated at 980 $^{\circ}$ C for 10 min, cooled to room temperature for 300 s, held in room temperature for 3 min and then cooled to -150 $^{\circ}$ C for 15 min, b) region of martensitic transformation in Fig 2a at a higher magnification.

at cryogenic temperature. In the interrupted cooling mode, when the steel cooled down from the ambient temperature, martensitic transformation did not start immediately. Under these conditions, there is a second M_s temperature (-21 °C) that is related to the re-start of transformation after stopping at the ambient temperature and provides the driving force to start the martensite transformation. The existence of the second M_s temperature in the martensitic transformation while cooling down to the cryogenic temperatures has been proved by other researchers in the case of D2 steel¹⁸. Regarding the low finish temperature of martensitic transformation, the use of cryogenic treatment to reduce or eliminate retained austenite in this steel is necessary.

The x-ray diffraction pattern of the studied samples is shown in Fig. 3. As can be seen, by performing cryogenic treatment the amount of retained austenite was reduced. The performed calculations using equation (1) showed that the retained austenite reached less than 4% in cryogenic treatment from 14.5% in conventional heat treatment. The reduction in the amount of retained austenite as the effect of the cryogenic treatment has also been proved on other steels¹⁴.

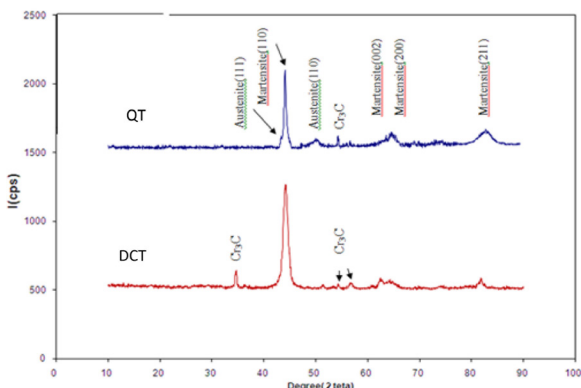


Fig. 3. XRD pattern of the INDRA5 steel: After quenching and after cryogenic heat treatment.

In Fig. 4, the optical microscope images of the quench-temper (Fig. 4a) and the quench-deep cryogenic-temper sample (Fig. 4b) are shown. As can be observed in Fig. 4b, fine carbides were produced in deep cryogenic treatment with a much better distribution compared to the carbides of the quench-temper sample. The calculations showed that carbide percentage from 5.7% in conventional heat treatment reached 7.9% in deep cryogenic treatment. The more appropriate distribution of carbides and their refinement due to deep cryogenic

treatment was also evident in the SEM image shown in Fig. 5. Two mechanisms are involved in cryogenic treatment: (1) transformation of the retained austenite to martensite and (2) precipitation of fine carbides. The reason for the precipitation of these carbides is that during cryogenic treatment, due to internal microscopic stresses caused by factors such as the difference in the thermal contraction coefficient of different phases and the transformation of retained austenite to martensite, crystalline defects such as twinning and dislocations were created. Under these conditions, if there is sufficient time, local diffusion will contribute to the clustering of carbon and alloy elements near these network defects. On the other hand, as the temperature decreased, super saturation increased in martensite, and thus the lattice distortion and thermodynamic instability increased in martensite. Both of the mentioned factors provided the driving force for carbon atoms and the alloying elements to move around the crystal defects, and caused the formation of very small nuclei. In tempering treatment (after cryogenic treatment), these nuclei lead to the creation of fine carbides in nanometer scale. The increase in volume fraction of these precipitations is the reason for the local diffusion of carbon atoms^{8, 16, 19}.

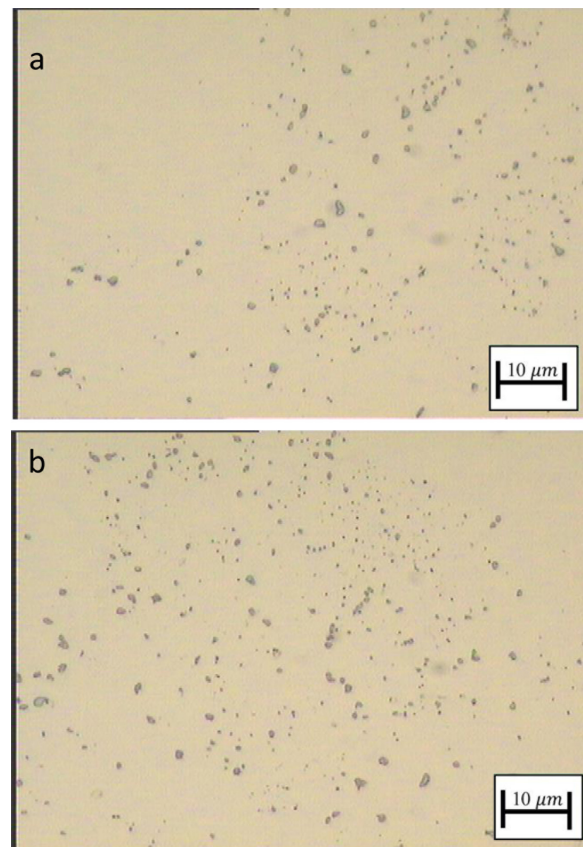


Fig. 4. Carbides distribution in the (a) QT, (b) DCT samples.

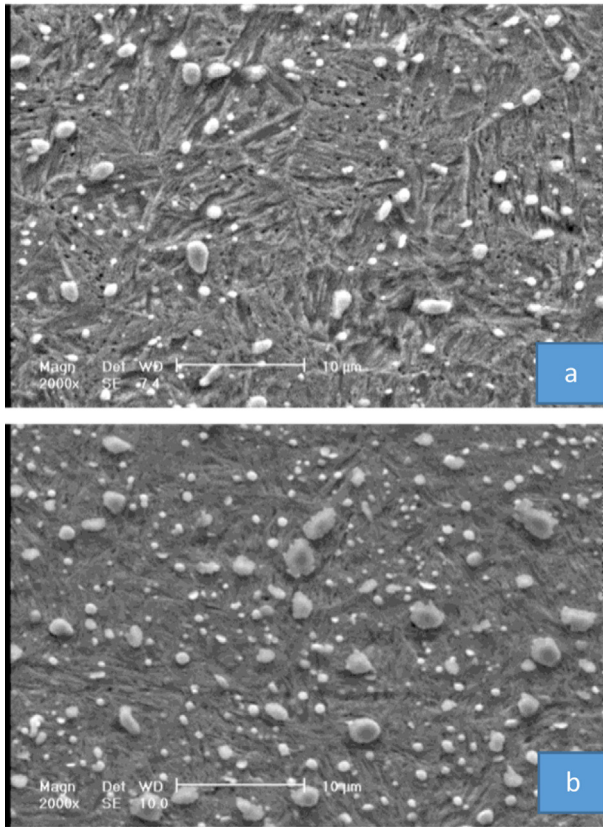


Fig. 5. SEM micrographs of the samples, a) conventional heat treatment (quench - temper) (QT), b) quench-deep cryogenic treatment for 24 hrs. – temper (DCT).

3. 2. Hardness and wear behavior

In Table 2, the hardness obtained from the cryogenic treatment and the quench-temper heat treatment is presented. The increasing hardness due to cryogenic treatment was related to the complete transformation of retained austenite to martensite and the precipitation of fine carbides and the increase in the volume fraction of these carbides. An increase in hardness due to cryogenic treatment has also been proved by other researchers ^{6,17}.

Table 2. Hardness of the samples after conventional and cryogenic treatments.

Type of heat treatment	Hardness (HRC)
Quench - Temper (QT)	60.5 ± 0.1
Quench- Deep Cryogenic Treatment in 24 hrs- Temper (DCT)	0.3 ± 63.2

In Fig. 6, the results of the pin- on- disk wear test are presented in form of a mass loss curve in terms of sliding distance under 120 N load and 0.2 m/s speed for the quench-temper and deep cryogenic-temper samples. It can be observed that both samples have a relatively constant decreasing rate of weight; however, lower weight loss rate in the cryogenic sample was quite clear. In Fig. 7, the relationship between the wear rate and the sliding distance in the wear test is presented. It was also observed here that the wear rate in the DCT sample was less than that of the QT sample. The improvement of wear behavior was estimated about 30%. In both samples, by increase of wear distance the wear rate decreased, which can be due to the increased work hardening of the samples. The main reason for the high wear resistance in DCT samples, in addition to the removal of retained austenite as a soft phase, is the increased percentage of alloy carbides due to deep cryogenic treatment and their more uniform distribution. The precipitation of fine carbides reduced the percentage of carbon and alloy elements in the matrix phase; therefore, the matrix toughness increased. Thus, it can be said that the sum of precipitation of fine hard carbides and an increase in the toughness of the matrix phase (martensitic) in the studied steel caused the reduction of the wear rate in the deep cryogenic treatment samples ⁸.

In order to determine the wear mechanism, the worn-out surface was investigated by SEM, the results of

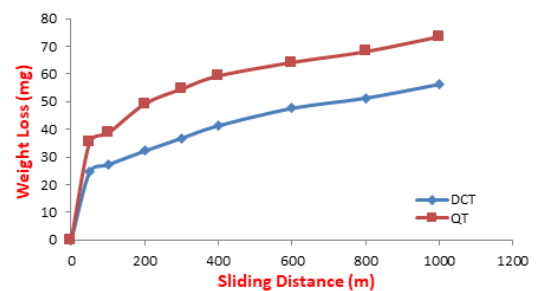


Fig. 6. The relationship between the sliding distance and weight loss under 120 N load and V=0.2 m/s.

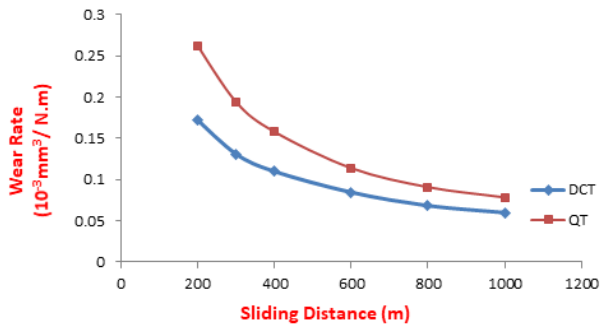


Fig. 7. The relationship between the sliding distance and wear rate under 120N load and $V=0.2$ m/s.

which are presented in Fig. 8. In this Figure, adhesive particles to the surface are observed, which indicate the dominance of adhesive wear mechanism in the studied samples. Due to the cryogenic treatment, the size and number of adhesive pieces to the surface (the downs and heights of the surface), which are in fact the products of adhesive wear, were reduced (Fig. 8a). This can be justified regarding the reduction in the wear rate in the cryogenic samples compared to the non-cryogenic ones (quench-temper heat treatment).

The chemical analysis performed on the worn-out surfaces (Table 3) showed that in the adhesive patches, there was also tribochemical wear due to high carbon

percentage in these patches. Therefore, the wear mechanism in the mentioned samples was as follows:

- In the conventional heat treatment sample: severe adhesive (adhesive multilayer patches) and tribochemical mechanisms;
- In the deep cryogenic sample: mild adhesive (scattered adhesive patches) and tribochemical mechanisms.

The investigation of the wear products in this steel by SEM (Fig. 9) illustrated the fragility of these products. Considering the better distribution of alloy carbides and the removal of retained austenite in deep cryogenic samples, the particles produced by wear in these samples were smaller than the samples under the quench-temper heat treatment. The fragmentation of the wear products in cryogenic treatment for other tool steels has been proved by other researchers²⁰⁾. In studies by Fantalou et al.²¹⁾ on tool steels, the increase in volume fraction and reduction in distance between carbides are mentioned to be the reasons for the reduction of adhesive wear. Likewise, adhesive wear has a direct proportionality to the applied force and reverse proportionality with hardness.

Yang et al.²²⁾ considered the increased hardness of the surface of steel to be the main factor in improving resistance against adhesive wear. Finally, regarding the above mentioned cases, in deep cryogenic treatments

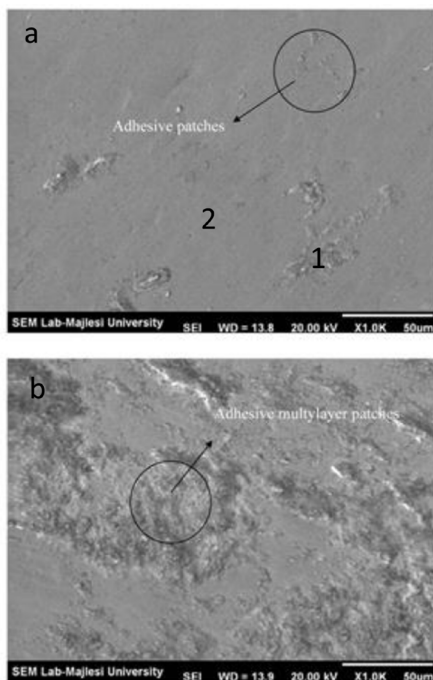


Fig. 8. SEM images of worn – out surfaces at high magnification, in pin-on-disk method, under 120 N load and $V=0.2$ m/s, a) quenched - deep cryogenic treatment for 24 h - tempered sample, b) quenched-tempered sample.

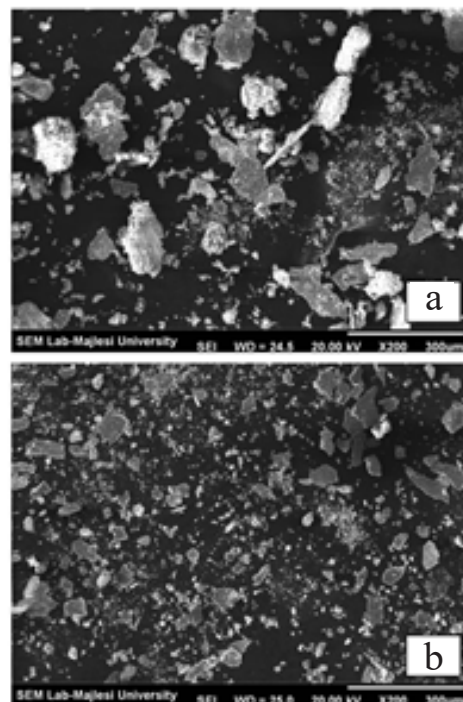


Fig. 9. Wear debris of a) quenched - tempered samples, b) quenched - deep cryogenic treatment for 24 h- tempered samples, under 120 N load and $V=0.2$ m/s.

Table 3. Chemical analysis of points 1 and 2 in Fig. 8a.

Fe%	Cr%	Mo%	Mn%	Si%	O%	
76.233	4.435	0.708	0.131	1.063	17.429	Point 1
92.330	4.373	0.134	0.253	0.874	2.036	Point 2

performed on the studied steel, the adhesive wear was reduced due to an increase in volume fraction of carbides and their fragmentation and eventually increased hardness (due to an increase in the percentage of martensite and precipitation of fine carbides).

4. Conclusion

- Regarding the results of the dilatometer test, the starting and finishing temperatures of the martensitic transformation in INDRA 5 steel in interrupted cooling were 234 °C and -102 °C, respectively. In addition, in the interrupted cooling mode, the martensitic transformation did not immediately start by cooling the steel from the ambient temperature. Under these conditions, there was a second Ms temperature (-21 °C) that was related to the re-start of the transformation after stopping at ambient temperature and providing the driving force to start the martensitic transformation.
- Applying deep cryogenic treatment on this steel caused a reduction in the amount of retained austenite and precipitation of fine carbides, an increase in volume fraction of carbides, and more appropriate distribution of these carbides. Thus, the hardness increased by 3 units (HRC scale) and the wear resistance increased 30%.
- The wear mechanisms in the quench-temper samples (conventional heat treatment) was severe adhesive together with tribochemical mechanisms which were converted to mild adhesive and tribochemical mechanisms due to the cryogenic treatment. Moreover, the collected wear debris of the cryogenically treated samples were more brittle and smaller.

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