Friction and Wear Behavior of Irradiated Polyethylene Sliding Against a Rough Steel Surface

W. Tuckart, E. Molinari, D. Rossit & M. Failla

Tribology Letters

ISSN 1023-8883

Tribol Lett
DOI 10.1007/s11249-014-0344-x

Springer

Tribol Lett
DOI 10.1007/s11249-014-0344-x

ORIGINAL PAPER

Friction and Wear Behavior of Irradiated Polyethylene Sliding Against a Rough Steel Surface

W. Tuckart · E. Molinari · D. Rossit · M. Failla

Received: 14 November 2013 / Accepted: 28 April 2014

Abstract The aim of this study was to evaluate the tribological behavior of polyethylene crosslinked by gamma radiation sliding against a steel surface. Two high-density polyethylenes were irradiated to a total dose in the range of 2−20 Mrad under vacuum and at room temperature. After irradiation, the materials were annealed at 423 K and then cooled slowly to room temperature. The same thermal treatment was applied to the non-irradiated polymer. The wear behavior of the polymers was determined under controlled ambient temperature of 298 and 333 K using a homemade tribometer. Sheet-shaped specimens were loaded against the surface of a steel disc with different normal loads to generate nominal contact pressures in the range of 0.25–1.5 MPa. The tests were performed under dry conditions using a disc rotation to produce an average sliding speed of 0.6 m/s and during a period of time to provide an average sliding distance of 1,080 m. The wear rate was obtained as the mass loss by the sample divided by the sliding distance, and the friction coefficient was determined by measuring the friction force. The results indicate that the wear rate increases with load in the case of non-irradiated polyethylene and low-dose irradiated polymers, while the wear rate reaches a maximum value with the load in the case of the irradiated samples with high doses. The samples irradiated with a dose of 10 Mrad showed the lowest wear. The coefficient of friction (COF) increases slightly with the load in all the cases. Most irradiated polymers show higher COF than the non-irradiated material when compared at a given load. The results show that the irradiation dose applied to the polyethylenes produced no noticeable effect on the COF values when a comparison was made at a given applied load.

Keywords Abrasive wear · Cross-linked · Polyethylene · Friction

1 Introduction

The use of polymeric components with contact surfaces in relative movement has spread widely in various branches of engineering where high performance is required in terms of tribological sliding and/or friction, because they combine relatively low cost with good resistance to wear and self-lubricating characteristics [1]. Ultra-high-molecular-weight polyethylene (UHMWPE) is among the polymeric materials known for their low coefficient of friction (COF) and excellent wear resistance, making it suitable for many applications. A great amount of work has been devoted to understanding and establishing the factors that affect the wear behavior of UHMWPE; in addition, various treatments have been developed in order to improve its wear resistance. Most of them are based on producing molecular crosslink [2–5]. Some authors found that crosslinking considerably decreases the abrasive and adhesive wear of UHMWPE when sliding over different materials [6, 7]. Crosslinking limits molecular orientation and enhances the flow resistance of the material at the surface, resulting in better resistance to wear [8, 9].

Despite its expanded use in tribological applications, a remaining disadvantage to using UHMWPE is its high melt

W. Tuckart (✉) · E. Molinari · D. Rossit · M. Failla
Grupo de Tribología, Departamento de Ingeniería, Universidad Nacional del Sur, CONICET Avd. Alem 1253, 1° (8000), Bahía Blanca, Argentina
e-mail: wtuckart@uns.edu.ar

M. Failla
Planta Piloto de Ingeniería Química (PLAPIQUI), UNS-CONICET, C.C. 717 (8000), Bahía Blanca, Argentina

Published online: 16 May 2014

viscosity, which hampers the use of conventional melt-processing techniques. Different strategies are followed in order to obtain melt-processible polyethylenes (PEs), which in turn have better wear resistance. Therefore, strategies to improve the wear resistance of the polymer include controlling its molecular weight and the width of the molecular weight distribution [10], blending small amounts of UHMWPE [11, 12], forming molecular crosslinking by irradiation with electron or gamma rays, or chemical attack with organic peroxide.

The majority of the publications on the influence of molecular crosslinking on the response to the friction and wear of polyethylene are concerned with UHMWPE. Few studies dealing with the tribological behavior of crosslinked regular PE have been reported [13–17]. Early investigations showed that under identical sliding wear conditions, crosslinked PE has greater wear resistance than the nonirradiated polymer if the gamma irradiation process is carried out in air atmosphere, while the irradiated PE has lower wear resistance than the original PE if the irradiation is performed under vacuum [13]. Matsubara and Watanabe [14] studied the friction and wear of gamma-irradiated high-density PE sliding on steel coated with chromium. A PE was irradiated under vacuum to a total dose ranging from 5 to 2,000 Mrad. The authors found that under nominal pressure of 1 MPa, the wear of the materials increases with increasing radiation doses up to a critical dose of 200 Mrad and then suddenly decreases at higher dose. The nonirradiated PE wear was lower than that for irradiated polyethylenes when wear tests were performed using normal pressures below 1 MPa; however, at higher pressures the wear of the nonirradiated PE increases sharply, reaching much higher values than that corresponding to the PE irradiated with doses above 100 Mrad. It was suggested that an important factor determining this phenomenon is the rise in the temperature of the sliding surface; when the temperature exceeds the melting point, the polymer reaches a rubber state having good wear resistance. We found in a previous study that the wear of irradiated high-density polyethylenes was similar to the corresponding nonirradiated material when sliding against steel under a nominal pressure lower than 1 MPa, while at higher contact pressure, the irradiated material wear was less than that of the unirradiated one [18]. The tribological studies conducted on gamma-irradiated UHMWPE are not conclusive regarding the effect of the irradiation on the wear resistance of the polymer. Some authors found that the wear resistance improves after irradiation [5], while others reported either a decrease or insignificant effect of irradiation on the wear of the material [9–19].

The difference observed in response to the wear of irradiated polyethylenes may be due to the combination of the various factors affecting the structure of the polymers with the fact that the tribological properties are not true material properties. These properties depend on a complex combination of factors such as the nature and topography of the metal contact, sliding speed, applied load and temperature, to name just a few. Gamma irradiation is known to produce free macro radicals in PE that can lead to chain linking. When the irradiation of the polymer is performed at relatively low temperature, i.e., room temperature, the molecular structure, crystallinity level, environment and post-irradiation treatment are among the most important factors affecting the crosslinking process. This in turn determines the characteristics of the end structure and the physical and mechanical properties of the material [2–4, 14, 20].

In the present work, we studied the friction and wear behavior of two gamma-irradiated linear high-density PE sliding against a rough steel surface. The polymers with different average molecular weights were irradiated under vacuum to total doses in the range from 2 to 20 Mrad. The irradiated materials were thermally treated at temperatures above the melting point to eliminate free radicals and reduce the chances for long-term oxidation. The proportion of gel produced was determined by solvent extraction. The microVicker hardness and enthalpy of fusion as measured by calorimetry were determined. The wear rate and COF were determined under dry sliding conditions at 298 and 333 K in air atmosphere. Thus, we can examine the influence of the initial molecular structure, dose and gel amount on the morphology and hardness and their relationship with the tribological behavior.

2 Experimental Procedure

2.1 Materials and Methods

Two high-density polyethylenes supplied by DuPont de Nemours and Oxy Petrochemical were used in this study. The polymers have an average molecular weight of 56,000 and 80,600 g/mol, which were identified as PE5 and PE8, respectively. The polydispersity was 2.6 for both polymers.

The PE sheets were prepared by compression molding at 423 K using a hydraulic press with thermostatically controlled platens. The samples were molded between 0.5-mm-thick steel plates held apart by 0.5-mm-thick brass spacers. Then, they were quenched from the melt to ice-water temperature.

Strips with 12 mm length, about 0.5 mm thickness and 10 mm width were cut from the films and inserted into glass tubes. The tubes were evacuated to 0.133 Pa for 2 days and then sealed off. Subsequently, these samples were exposed, at room temperature, to γ-rays generated by a ^{60}Co source at a Comision Nacional de Energía

Atómica (CNEA-CAE) facility. The dose rate was 0.33 Mrad/h determined by dosimetry with a radiochromic thin-film dosimeter. Equal doses of 2, 5, 10 and 20 Mrad were applied to the samples. After irradiation, the samples were thermally treated at 423 K for 4 h while still inside the glass tubes to reduce the concentration of surviving free radicals and thus to avoid long-term oxidation. After the thermal treatment, the samples were allowed to cool down slowly to room temperature and then exposed to air. The nonirradiated polyethylenes were subjected to a thermal process equivalent to that applied to the irradiated samples. The irradiated samples are identified with the code PEx-# where x and # identify the polymer and the applied dose, respectively; for example, PE8-5 identifies the polymer PE8 that received a total dose of 5 Mrad.

The gel fraction of the irradiated samples was determined by extraction of the soluble portion using xylene at 398 K, and the level of crystallinity of the samples was determined from the enthalpy of fusion, which was measured in a Perkin-Elmer Pyris I DSC using a heating rate of 10 K/min. The heat of fusion (ΔH_f) was obtained from the area of the endothermic peak, which was used to estimate the degree of crystallinity of the samples. The crystallinty was calculated by assuming the heat of fusion of 100 % crystalline polyethylene (ΔH_f^0) to be 69 cal/g [21]. The percentage crystallinity is then calculated as follows: % crystallinity $= (\Delta H_f/\Delta H_f^0) \times 100$ %.

The hardness of the samples was obtained from Vickers microhardness tests performed at room temperature. A load of 0.40 N was applied on the indenter and then released after 2 min. The length of the projected diagonals of the indentation mark was measured with the help of an optical microscope (OM). The Vickers pyramidal number was calculated from

$$\mathrm{VH} = 2P\sin(\alpha/2)/d^2$$

where P is the applied load, α is the angle of the pyramid (136°), and d is the average length of the diagonals of the indentation mark in mm.

2.2 Sliding Wear Test

A homemade tribometer equipped with a temperature-controlled environmental chamber was used to assess the behavior of materials subjected to unidirectional sliding against steel without using lubrication. Figure 1 displays the schematic side view (a) and top view (b) of the flat-on-disk type assembly showing the arrangement of the sample holder, sample and steel disc.

Sheets of the polymers with a size of 10 × 10 mm were loaded against the surface of a steel disc of 40 mm diameter turning at 375 rpm. The radial distance over which the sample contacted the axial surface of the disc was about 10 mm as measured from the outer edge of the disc. The average sliding speed was 0.6 m/s, which was calculated at a disc diameter of 32 mm, and the tests were run for a period to provide the average sliding distance of 1,080 m. The normal load on the sample was generated by a dead weight. A load of 2.5, 5, 7.5 or 10 N was applied to the sample when testing PE5 and PE5-# materials, while a load of 7.5, 10, 12.5 or 15 N was used when testing PE8 and PE8-# materials. The environmental temperature was controlled at either 298 ± 1 or 333 ± 1 K.

The mass lost by the samples after the wear test was measured using a precision balance with sensitivity of 10^{-4} g. The wear rate was obtained as the mass loss divided by the sliding distance; the wear mean value reported was obtained by averaging the results of at least three tests for each condition. The friction force was measured with a load cell and continuously recorded with a data acquisition system. The COF was obtained from the mean friction force calculated from the force trace after the initial sliding

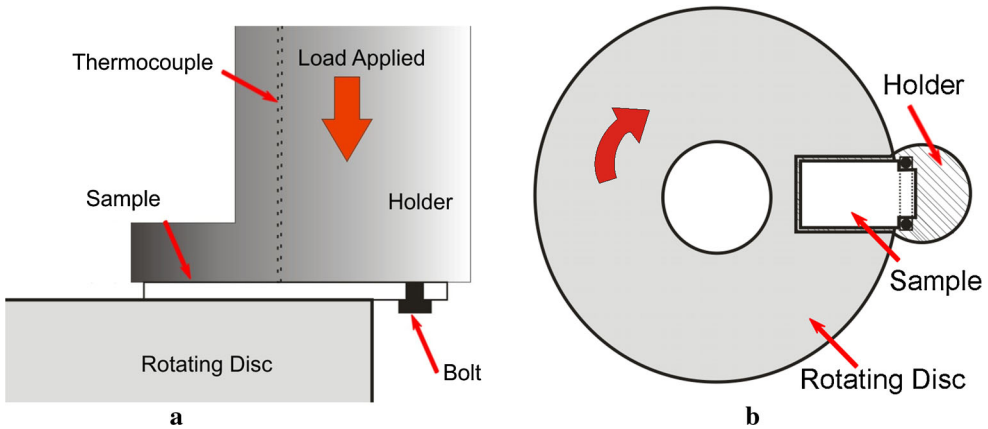

Fig. 1 Diagram illustrating the assembly formed by the sample holder, sample and tsteel disc. **a** *Lateral* view and **b** *upper* view

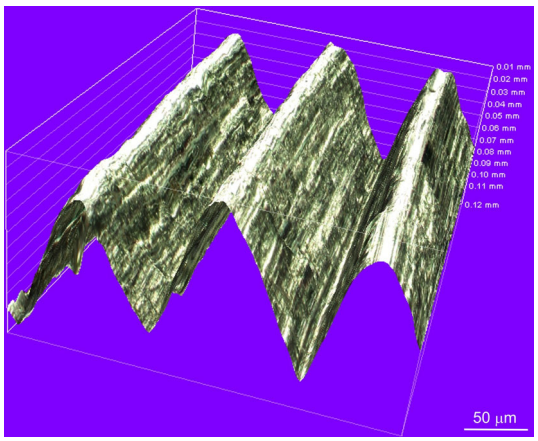

Fig. 2 3D view of the topography of the disc surface

Table 1 The amount of gel and crystallinity level, both expressed as percentage, and the hardness of the materials

Material	Dose (Mrad)	Gel (%)	Crystallinity (%)	Hardness (kg/mm^2)
PE5	0	0	70	5.2 ± 0.1
	5	9	60	5.1 ± 0.2
	10	43	56	3.6 ± 0.2
	20	67	54	3.9 ± 0.1
PE8	0	0	71	4.6 ± 0.6
	2	0	64	4.1 ± 0.1
	5	38	59	5.2 ± 0.5
	10	76	57	4.2 ± 0.2
	20	85	56	4.5 ± 0.1

period in accordance with ASTM standard G115-04. The deviation in the value of COF was about ±20 %.

Discs of 5 mm thickness of SAE 4140 steel were obtained using a lathe machine capable of providing rough surfaces to cause wear of the polymer mostly by the mechanism of abrasion [23]. Figure 2 displays the surface characteristics of the disc where peaks and troughs produced by the machining process are noticeable.

The roughness of the surface along the direction of the radius of the disc was measured according to ISO 3274:1996 using a Hommel T500 roughness meter and Etamic software. The roughness parameters measured were $R_a = 24 \pm 0.01$ µm, $R_t = 167 \pm 0.9$ µm and $R_z = 162 \pm 0.44$ µm. R_t is the difference between the height of the highest point and the lowest point within the distance of a profile point sampling; R_z is the difference between the average height of the five highest points and the average height of the five lowest points of the profile point.

The contact surface of the disc and the polymer after the sliding test was examined with an OM and scanning electron microscope (SEM). The polymer samples were sputter-coated with a thin layer of gold prior to SEM examination.

3 Results

The amount of gel developed upon irradiation and the crystallinity levels of the samples after the thermal treatment applied to the samples are given in Table 1. At low dose, the effect of radiation is to increase the average molecular weight due to chain linking. No formation of a macroscopic gel in the case of PE8-2 was found, but this sample has an average molecular weight larger than the original PE8 [22]. At higher dosage, an insoluble gel forms that increases with the dose. These results are expected, and they are in concordance with those found by other authors when studying polyethylene irradiated with high-energy ionizing radiation under vacuum [23–25].

Table 1 also shows that the irradiation dose does not seem to affect the hardness of PE8 materials, while the materials obtained by irradiating PE5 with higher doses have hardness lower than PE5. This result may be associated with the competing effect of the reduction in the crystalline level and the increases of the chain linking level with the dose. It was observed that the microhardness increases progressively with increasing doses in irradiated PEs [26], while this property shows an increasing trend with the crystallinity in non-irradiated PEs [27].

Table 2 includes the wear data results obtained at the temperatures studied. Figure 3 shows the wear rate as a function of the load after testing the materials at 333 K. The figure shows that the wear of PE5 is relatively low, increasing almost proportionally with the load up to 7.5 N. Above 7.5 N, the wear seems to increase rapidly. The wear of PE5 was extremely large when loads above 10 N were applied, producing complete destruction of the sample. The wear of PE8 increases proportionally with the load in the range of loads studied. When the data results for the non-irradiated materials are compared at the same applied load, PE8 shows higher wear resistance than PE5. The difference in the wear between the polymers becomes larger as the load increases. For example, when loads of 7.5 and 10 N are applied, wear of PE5 is respectively 1.5 and 4 times that of PE8. The results in Table 2 indicate that the temperature does not produce a clear effect on the wear of PE5. At a load of 2.5 N, the wear of the polymer increases with temperature, while for loads of 5 N and 7.5 N, the wear diminishes with increasing temperature. At 10 N, which was the highest load applied to PE5, the increment in

Table 2 Wear rate of the material ($\times 10^{-7}$ g/m)

Material	Dose (Mrad)	Temperature (K)/normal load (N)							
		298/2.5	333/2.5	298/5	333/5	298/7.5	333/7.5	298/10	333/10
PE5	0	1.1 ± 0.04	3.7 ± 0.4	13.2 ± 1.6	10.1 ± 2.0	22.8 ± 3.2	15.4 ± 6.0	58.9 ± 6.0	59.4 ± 4.2
PE5-5	5	1.6 ± 0.2	0.5 ± 0.01	4.8 ± 0.7	10.6 ± 3.0	18.5 ± 8.0	21.1 ± 9.0	11.1 ± 2.7	37.1 ± 9.0
PE5-10	10	0.5 ± 0.03	1.6 ± 0.03	3.5 ± 0.8	11.2 ± 2.7	7.40 ± 0.5	14.5 ± 3.2	13.5 ± 1.4	11.2 ± 3.3
PE5-20	20	0.5 ± 0.02	0.5 ± 0.12	5.3 ± 0.6	5.3 ± 0.5	27.6 ± 13	23.0 ± 2.0	22.2 ± 11.1	26.0 ± 9.0
		Temperature (K)/normal load (N)							
		298/7.5	333/7.5	298/10	333/10	298/12.5	333/12.5	298/15	333/15
PE8	0	4.3 ± 0.1	9.8 ± 2.9	6.9 ± 0.1	13.0 ± 6.0	5.8 ± 1.5	19.1 ± 6.7	30.3 ± 10.0	24.4 ± 0.5
PE8-2	2	1.1 ± 0.3	6.4 ± 3.8	7.4 ± 0.8	18.0 ± 0.5	8.7 ± 0.1	19.6 ± 2.2	19.1 ± 6.9	17.5 ± 3.1
PE8-5	5	1.6 ± 0.4	9.0 ± 1.6	4.8 ± 0.5	11.7 ± 0.3	1.1 ± 0.1	11.7 ± 3.3	22.8 ± 3.2	18.7 ± 0.9
PE8-10	10	1.9 ± 0.3	1.6 ± 0.3	2.1 ± 0.1	5.8 ± 0.07	2.6 ± 0.3	3.7 ± 0.3	5.3 ± 2.6	8.2 ± 2.6
PE8-20	20	2.6 ± 0.03	5.3 ± 0.3	2.4 ± 0.03	7.4 ± 1.5	23.4 ± 3.0	13.2 ± 2.5	7.2 ± 0.3	–

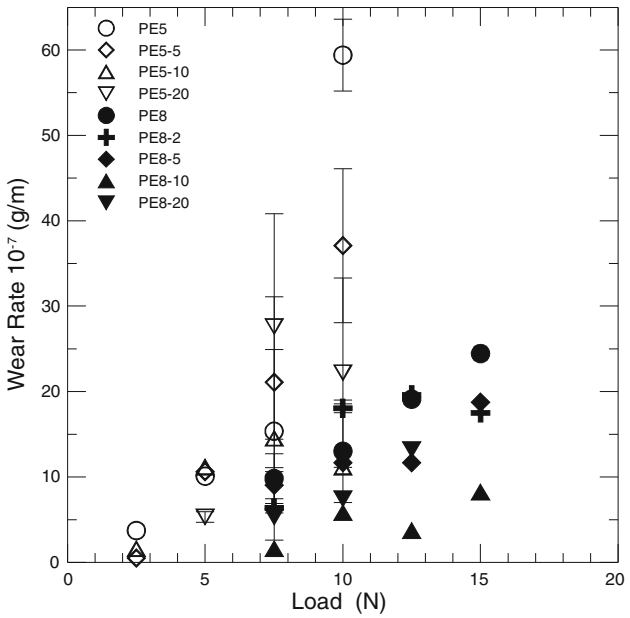

Fig. 3 The wear rate as a function of the load at 333 K

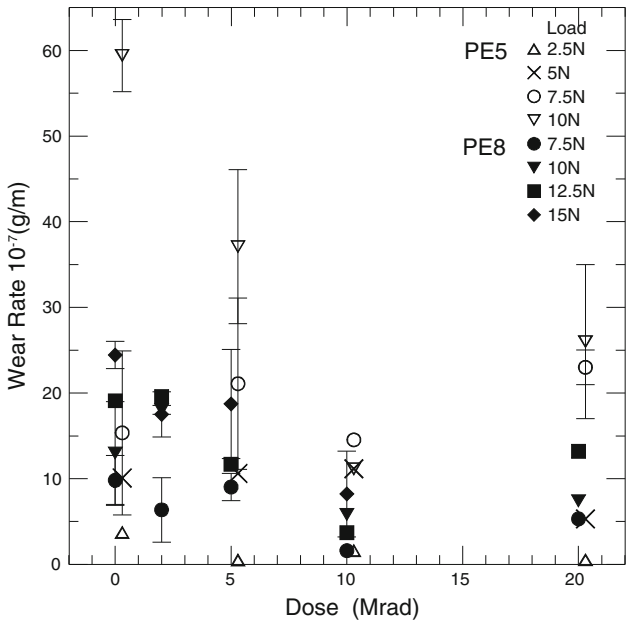

Fig. 4 The wear rate as a function of the dose at 333 K

temperature does not seem to affect the wear of the polymer. PE8 wear appears to increase with the temperature when comparing the data at a given load, except for the 15 N load, in that the wear decreases to some extent with increasing temperature.

Figure 3 shows that the wear of the irradiated material shows a trend of increasing with loading. At a given load, the wear of the polymers obtained by irradiating PE5 is comparable to, or even lower than, that corresponding to PE5, but for PE5-5 and PE5-20 tested at a load of 7.5 N, they show higher wear than PE5. Figure 3 also shows that at a given load most irradiated PE8 wears less than non-irradiated PE8, but PE8-2 shows greater wear than PE8 at a load of 10 N. Analyzing the wear results at 298 K shown in Table 2, it can be seen that, within the dispersion that exists in the data values, the majority of the irradiated material has lower wear than the original PE; one exception is PE8-20, which shows greater wear than PE8 when a load of 12.5 N is used.

The results in Table 2 indicate that the temperature has no specific influence on the wear of the irradiated material obtained from PE5. At a given load, and depending on the

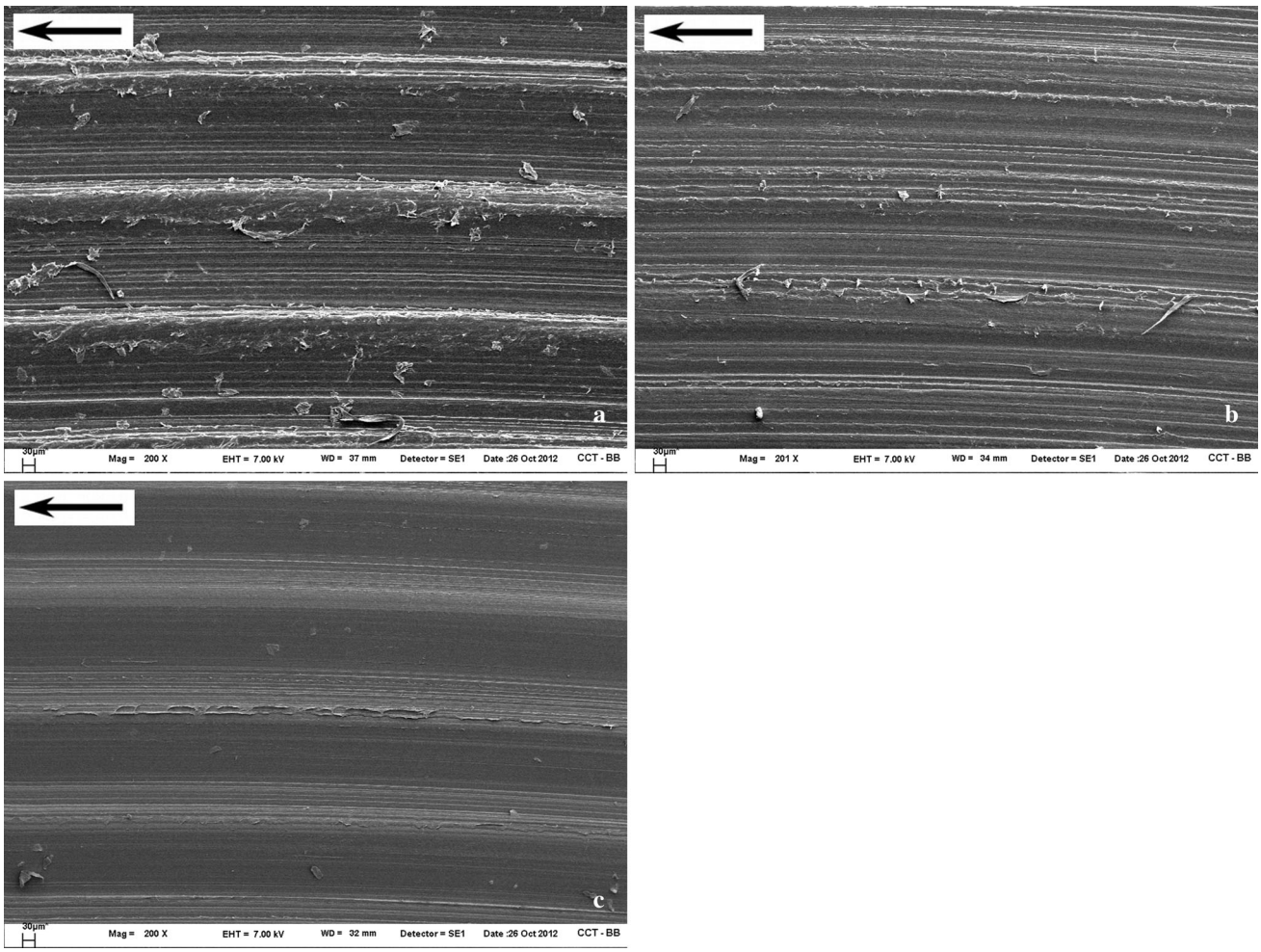

Fig. 5 SEM micrographs of the worn surface produced after testing PE8 **a**, PE8-5 **b** and PE8-10 **c**. Sliding direction *parallel* to the *horizontal*. Sliding conditions: 15 N and 298 K

material, the wear may increase or decrease, or not significantly change, with increasing temperature. The wear of irradiated PE8 materials seems to increase with the temperature. However, PE8-2 and PE8-5 show a small decrease in wear with the temperature when tested with a load of 15 N.

As an illustrative example of the effect of dose on the wear rate, the data obtained at 333 K are displayed in Fig. 4, where the load is taken as the parameter. Figure 4 shows that the wear of the irradiated materials remains nearly constant with the dosage when the load used is equal to or lower than 7.5 N. The evolution of the wear with the dose seems to reach a limiting value or even to pass through a minimum localized at 10 Mrad in the irradiated material of both PEs that were tested with loads higher than 7.5 N. A qualitatively similar graph to that in Fig. 4 could be obtained if the wear measured at 298 K were represented as a function of dose. As seen in Table 2, the wear at 298 K seems to pass through a minimum value with the dose. The samples irradiated to a total dose of 5 or 10 Mrad present the lowest wear at a fixed load.

The characteristic of the surface of the polymer after sliding was examined by SEM microscopy. Figure 5 shows images of worn surfaces of PE8, PE8-5 and PE8-10, tested at 15 N, that were chosen as illustrative examples. On the surfaces of the materials, deep parallel grooves can be seen due to the grooving effect of the asperities of the steel counterpart. The damaged surface of PE8 (Fig. 5a) shows grooves, scratches and signs of plastic deformation of the material with the formation of strands, which are typically promoted by cohesive wear [28]. The sliding surface of the PE8-5 sample displayed in Fig. 5b is rather smooth with scratches with some evidence of plastic deformation of the material. The samples irradiated with doses of 2 and 5 Mrad showed wear surfaces with similar characteristics to that observed in Fig. 5b. The tribosurface of PE8-10 displayed in Fig. 5c is smooth with scratches promoted by abrasion wear. These surface characteristics were generally

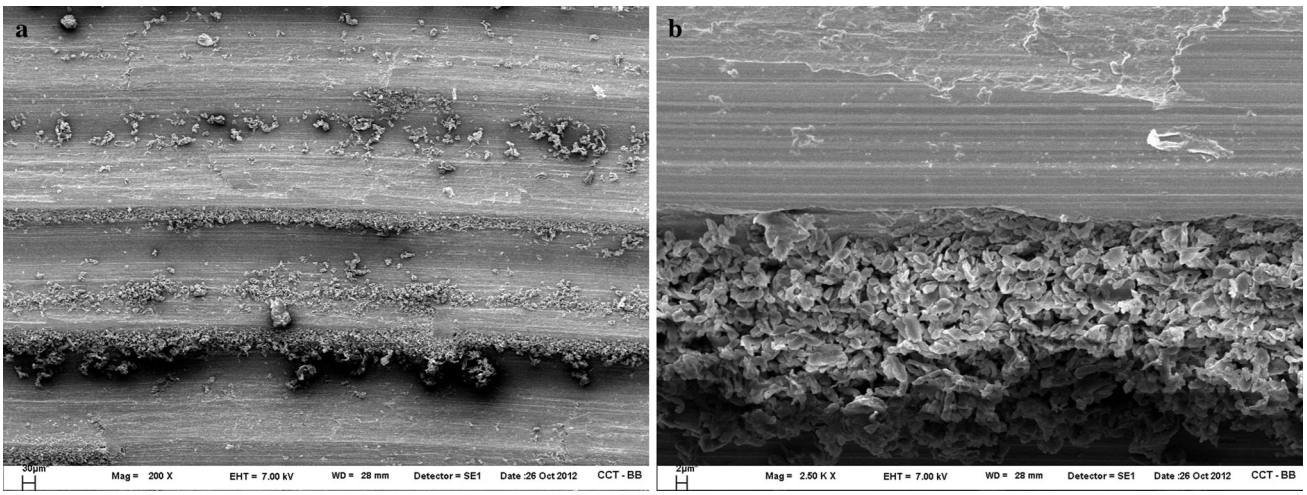

Fig. 6 SEM micrograph of the disc counterface after testing PE8-10 applying a load of 15 N

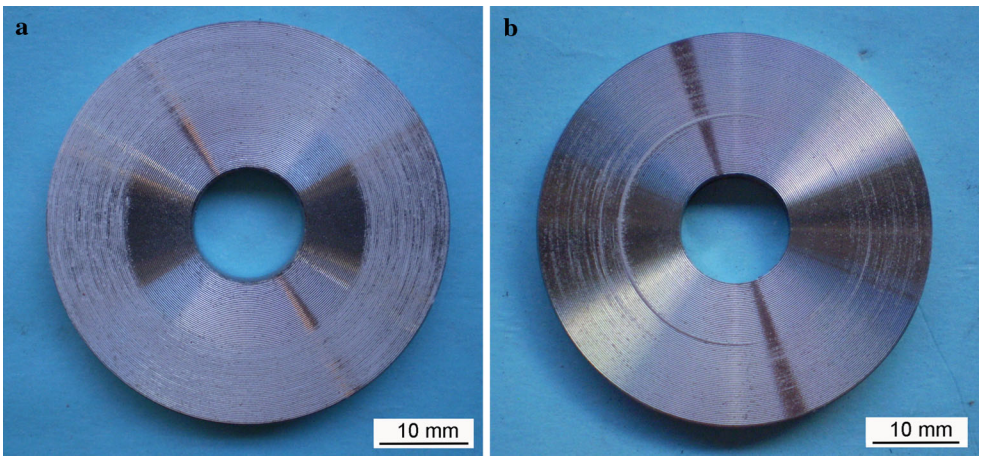

Fig. 7 Photographs of the steel disc after testing PE5 (**a**) and PE5-5 (**b**). Sliding conditions: 298 K and 5 N

observed in materials irradiated to doses of 10 and 20 Mrad.

Figure 6 includes SEM micrographs showing a top view of the surface of a disc, selected to illustrate the characteristic of the metallic surface commonly observed after testing the materials. Inspection of the surface in Fig. 6a shows fragments produced by the abrasion process, which have the appearance of fine cutting chips; they agglomerate and localize onto the slope surface and furrows of the metal grooves during repetitive sliding. Furthermore, it is evident that a transfer film of the material with patchy characteristics covers the steel surface, suggesting that adhesive processes occur. Figure 6b shows the surface of the disc at a high magnification where some polymeric material left over in the asperities is noticeable.

Figure 7 shows optical micrographs of the disc after testing PE5 (Fig. 7a) and PE5-5 (Fig. 7b) at 298 K using a normal load of 5 N. A heavy deposit of transferred material was noticed on the sliding track when PE5 was tested. While the deposited material on the sliding track (see Fig. 7b) seems to be lower than in the previous case, fragments of material gather at the edge of the wear track and appear uniformly distributed around the circumference.

The COF measured at different temperatures and loading is presented in Table 3 and plotted against load in Fig. 8. The COF of PE5 and PE8 measured at 298 K increases from 0.1 to 0.23 and from 0.08 to 0.2, respectively, following a trend almost proportional to the load. At 333 K, the COF of PE5 does not change significantly with load, while the COF of PE8 appears to increase slightly with load. The increment of the test temperature seems to decrease somewhat in the COF of PE5 and PE8 when they are tested with greater normal load.

The COF of the irradiated materials also showed some tendency to increase with the load at both analyzed

Table 3 Coefficient of friction of the materials

Material	Dose (Mrad)	Temperature (K)/normal load (N)							
		298/2.5	333/2.5	298/5	333/5	298/7.5	333/7.5	298/10	333/10
PE5	0	0.11 ± 0.01	0.10 ± 0.02	0.14 ± 0.01	0.13 ± 0.05	0.23 ± 0.01	0.11 ± 0.01	0.23 ± 0.01	0.13 ± 0.01
PE5-5	5	0.16 ± 0.02	0.10 ± 0.06	0.13 ± 0.01	0.18 ± 0.01	0.13 ± 0.01	0.20 ± 0.01	0.15 ± 0.01	0.21 ± 0.02
PE5-10	10	0.08 ± 0.01	0.13 ± 0.04	0.15 ± 0.005	0.17 ± 0.003	0.14 ± 0.01	0.24 ± 0.01	0.18 ± 0.02	0.16 ± 0.01
PE5-20	20	0.10 ± 0.03	0.12 ± 0.04	0.16 ± 0.01	0.16 ± 0.08	0.20 ± 0.02	0.16 ± 0.01	0.21 ± 0.02	0.19 ± 0.04
		Temperature (K)/normal load (N)							
		298/7.5	333/7.5	298/10	333/10	298/12.5	333/12.5	298/15	333/15
PE8	0	0.08 ± 0.01	0.11 ± 0.04	0.10 ± 0.02	0.11 ± 0.02	0.14 ± 0.01	0.17 ± 0.01	0.2 ± 0.01	0.15 ± 0.002
PE8-2	2	0.08 ± 0.01	0.13 ± 0.03	0.11 ± 0.04	0.15 ± 0.04	0.16 ± 0.01	0.16 ± 0.02	0.18 ± 0.01	0.13 ± 0.01
PE8-5	5	0.08 ± 0.01	0.16 ± 0.001	0.12 ± 0.01	0.15 ± 0.01	0.14 ± 0.01	0.16 ± 0.09	0.23 ± 0.02	0.14 ± 0.03
PE8-10	10	0.16 ± 0.02	0.17 ± 0.003	0.11 ± 0.02	0.16 ± 0.02	0.18 ± 0.03	0.21 ± 0.01	0.18 ± 0.02	0.15 ± 0.03
PE8-20	20	0.10 ± 0.01	0.13 ± 0.033	0.22 ± 0.05	0.21 ± 0.018	0.24 ± 0.005	0.19 ± 0.02	0.25 ± 0.01	0.21 ± 0.03

temperatures. In the case of the irradiated material, the increment in temperature from 298 to 333 K does not seem to affect the COF. However, at 333 K, the irradiated material displays a COF larger than those corresponding to the non-irradiated material. When the COF values of the irradiated materials are compared at a given load, it is not possible to establish a relationship between the COF and the dose received by each material for either tested temperature.

4 Discussion

4.1 Wear Sliding Behavior

In the present work, the wear behavior of HDPE and of the corresponding material crosslinked with various doses of gamma radiation was studied. As expected from the testing conditions used, there is evidence that abrasion is the main phenomenon that occurs in all the case [29]. The analysis of the tribosurfaces shows scratches, grooves and marks on the worn polymer surface, and the debris on the disc surface appears as fine cutting chips. Additionally, we also observed that there are signs of plastic deformation of the polymer in the surface region of the contact, and transfer of polymer occurs onto the steel surface, which is characteristic of adhesive wear [29]. The results are in concordance with those found by other authors who studied the wear of high-density polyethylene sliding against rough steel surfaces or other metallic surfaces under comparable conditions [30–33].

The results in Table 2 and Fig. 3 show that the wear rate of PE5 is almost proportional to the load up to 7.5 N after the rapid increment of the wear occurs. This wear behavior was observed for various polymers and for polyethylene in particular [5, 14, 33]. It was found that above a given combination of load and sliding speed, the wear rate of polyethylene considerably increases. This is associated with the influence of the frictional heating that can promote a temperature increase and softening of the polymer at the contact surface, causing severe wear, and the partial removal of the transfer material from the disc surface favoring the interaction between the polymer and the rough steel surface [14, 33, 34].

The wear rate of PE8 increases almost proportionally with the load in the load range studied, which is characteristic of abrasive wear. In this case, it seems that the load applied in the test was lower than that required to observe a change from mild to severe wear.

A comparative analysis of the PE's wear at a given load reveals that PE5 shows less wear resistance than PE8. These results are expected because of the difference between the average molecular weight of the polymer, PE5 being the one with the lower molecular weight. Other authors [10, 35] have already reported a decrease in the wear rate with increasing molecular weight of polyethylene. Tervoort et al. [10] proposed that an effective number of physical crosslinks per molecule is the main factor that determines the abrasive wear resistance of PE. The differences between the wear resistance of the PEs may also be explained considering their mechanical response to tensile forces and the simple theory of abrasive wear proposed by Lancaster, even though this has been verified for single transverse sliding conditions, which was not the case analyzed here [36–38]. The theory predicts that the wear rate is proportional to the applied load and the COF and inversely proportional to indentation hardness and toughness; the last property is usually estimated as the product of the tensile strength and elongation at break. The results

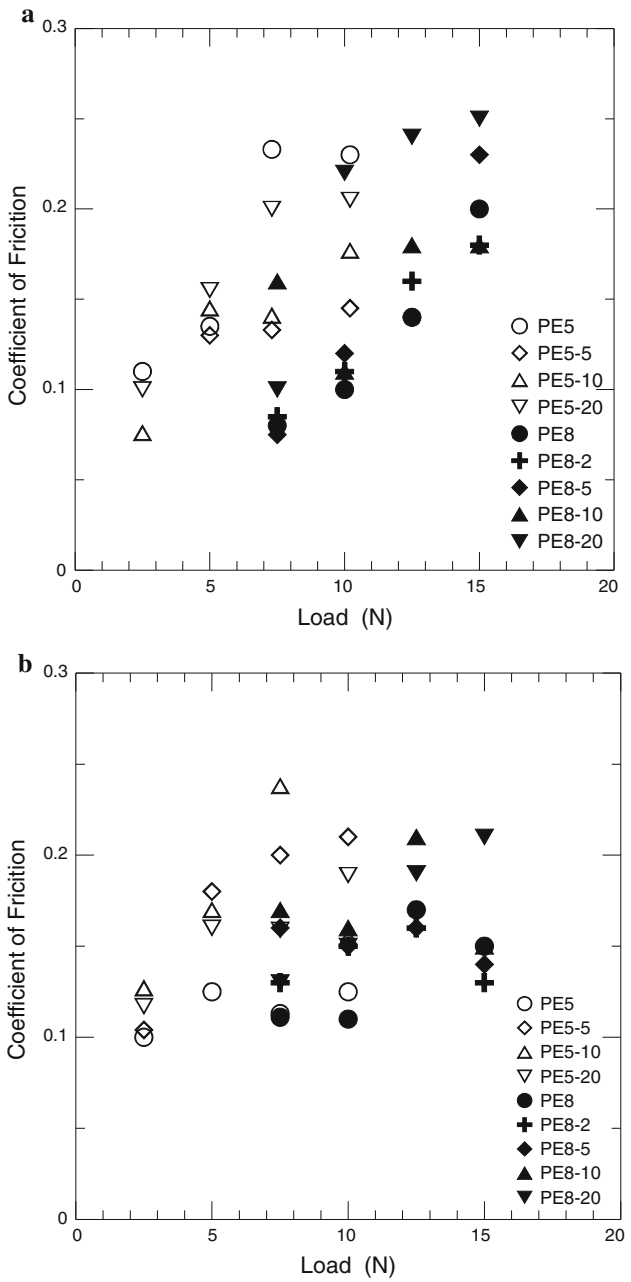

Fig. 8 Coefficient of friction as a function of the applied load: **a** 298 K and **b** 333 K

presented in Table 1 show that PE8 and PE5 have similar hardness, while Table 2 shows that the COF of PE8 is equal to or slightly lower than that of PE5, when compared at a given load. In addition, in previous works we studied the tensile response of the polyethylene and the irradiated materials in the temperature range from 298 to 383 K using typical tensile test conditions, which means that the conditions, i.e., deformation rate and stress field, are far apart from those that may exist at the contact sliding surfaces. It was found that PE8 behaved in a ductile manner, and the polymer showed strain hardening phenomena withstanding relatively large deformation before the break. Whereas PE5 displayed brittle behavior, the rupture occurs at quite low deformation and stress levels [22]. Therefore, the distinctive response of PE5 and PE8 to tensile force observed previously in combination with the results presented here may explain the differences that exist between the wear resistances of the polymers.

It was not possible to establish a relationship between the wear and the ambient temperature for each material. For polyethylene, an increment in the wear rate with the temperature is expected [40]. The lack of a connection between wear and ambient temperature may be attributed to the effect of both the rise in temperature at the contact surface and the accumulation of the debris and material transferred onto the surface of the disc. In the first case, the friction dissipation of energy may increase the interface temperature above the ambient one, reaching similar values in the test performed at a given load. In addition, the deposit of material onto the disc surface may lead to roughness changes and produce a lubricating effect, which hide the expected effect of the temperature on the wear.

The results presented in Table 2 and Fig. 3 show that irradiated polyethylenes may have similar or lower wear rates than the non-irradiated material, depending on at which load the comparison is made. In the range of low load, the wear of the irradiated and non-irradiated material is similar, while at the highest load used, the irradiated material displayed a lower wear rate than the non-irradiated material. The results differ somewhat from those found by Matsubara and Watanabe [14], who observed that the irradiated PE has lower wear resistance than the non-irradiated polymer when the sliding tests are performed using normal loads below the critical load required to observe the change from mild to severe wear. According to the results shown in Fig. 3 and Table 2, the wear rate of PE5 increases sharply as the load goes from 7.5 to 10 N. This suggests that, in this case, it may have reached the critical load. Such an increment in wear rate was not observed in the case of PE8, which indicates that larger load would be required to observe it.

Most of the irradiated materials display high wear resistance compared to the non-irradiated material when they are tested with loads above 7.5 N, the material irradiated with 10 Mrad being the one displaying the lower wear rate. This can be associated to the tensile mechanical response observed in the materials. In a previous study [39], we found that the elongation at break decreases continually with the dose, while the ultimate tensile stress increases, reaching a limiting value when the dosage is 10 Mrad. Therefore, considering the simple abrasive theory, the lowest wear observed in the case of the polyethylene irradiated to a total dose of 10 Mrad could be linked

to the mechanical properties. We tried to relate the tensile mechanical properties with the wear of the irradiated material, but the correlation was rather poor and therefore not included here [40].

We also tried to establish a correlation between the wear rate and gel or crystallinity level. The relationship between the wear and gel was analogous to that found with the dose owing to the connection that exists between the gel and dose. A representation of wear as a function of gel displays a minimum at gel of 40 wt% for the irradiated PE5 materials tested with the greater loads, while the minimum can be observed at gel of 75 wt% in the case of the irradiated PE8 materials.

The results in Table 3 seem to indicate that wear is not related to the initial crystallinity level of the polymers. This may be, at least partially, due to the plastic deformation of the material that appears to occur near the interface as well as the temperature increase that may occur in the contact area, which can alter the structure of the polymer near the surface. The results differ from those found by other authors that point out a decreasing wear rate with the crystallinity of the polymers [41, 42].

4.2 Friction Behavior

The average friction coefficient values obtained for all materials after sliding against steel under unlubricated conditions are presented in Table 3 and Fig. 8. The COF values fall in the range of 0.1 to 0.23, which are coherent with those reported for polyethylene sliding against steel, although it is necessary to consider that the friction results are affected by material transfer to the steel surface during repetitive sliding [5, 33, 43–45].

Figure 7a shows that the COF of PE5 and PE8 increases with the load at 298 K. The results go along with those presented by several authors where the friction coefficient increases with the load, which might be explained by the contribution of plastic deformation to the friction force [29, 34, 46]. As was discussed above, PE5 wears more than PE8. In general, the COF of the irradiated material measured at 298 K increases with load. The COF of the irradiated material is lower than those reported by Matsubara and Watanabe [14] for irradiated polyethylene; they found COF values in the range of 0.3–0.4, but these values were collected from tribotests made in the range between 100 and 400 N of normal load.

The results measured at 333 K in Fig. 8b show that the COF value remains practically constant as the load increases in the case of PE5, meaning that the friction and normal forces are proportional. Meantime, the COF of PE8 increases just slightly with the load. Increasing the test temperature from 298 to 333 K seems to affect the value of the COF significantly when larger loads are applied to both polyethylenes. The changes in the COF may be linked to the decrease in the mechanical properties, elastic modulus and strength of the polymers with increasing temperature [39]. In the case of the irradiated material, the increment in the temperature from 298 to 333 K does not seem to affect the COF. However, at 333 K, the irradiated material displays COFs larger than those corresponding to the non-irradiated material. The polymer transferred to the steel surface might explain the differences in the COF values between non-irradiated and irradiated materials. The examination of the steel surface after the test showed that the wear process of the non-irradiated PE produces an appreciable deposit of material covering most of the sliding track. However, the amount of transferred material was significantly lower in the case of the irradiated material, which may increase the contact between the polymer and steel surfaces affecting the COF.

The results show that the irradiation dose applied to the polyethylenes produce no noticeable effect on the values of COF when a comparison is done at a given applied load.

5 Conclusions

The study of the wear and friction of high-density polyethylenes and irradiated polymers sliding against a rough steel surface shows the following:

- The polymer PE8 displays higher wear resistance than PE5 when compared at a given normal load. This may reflect the influence of the molecular weight and the mechanical properties of the polymer on its wear performance.
- It is likely that the main wear mechanism was abrasion; however, evidence exists that an adhesion component may also contribute.
- The irradiation dose applied to the polyethylenes does not noticeably affect the wear of the polymers as long as the normal load used in the sliding test is lower than 10 and 12.5 N in the case of irradiated PE5 and PE8, respectively. The PE5 and PE8 irradiated with doses of 5 and 10 Mrad, respectively, show the lowest wear when the tribological test is carried out using the highest normal load.
- The COF of the nonirradiated polyethylenes increases slightly with the normal load at 298 K, while the COF of these materials does not vary significantly with the load when the sliding test is done at 333 K.
- The COF of the irradiated material increases slightly with the load at both tested temperatures. At 333 K, the COF of the irradiated polyethylene is mostly higher than that of the original polymer when they are compared at a given load.

Acknowledgments The authors wish to express their appreciation for the support given to the Engineering Department of Universidad Nacional del Sur and CONICET for the financial support of this article.

References

1. Brostow, W., Corneliussen, R.: Friction and Wear of Failure of Plastics. Hanser, New York (1986)
2. Lyons, B., Weir, F.: The Radiation Chemistry of Macromolecules. Academic Press, New York (1973)
3. Chodák, I.: Properties of crosslinked polyolefin-based materials. Prog. Polym. Sci. **20**(6), 1165–1199 (1995)
4. Failla, M., Vallés, E., Lyons, B.: Effect of initial crystallinity on the response of high-density polyethylene to high-energy radiation. J. Appl. Polym. Sci. **71**, 1375–1384 (1999)
5. Shen, C., Dumbleton, J.H.: The friction and wear behavior of irradiated very high molecular weight polyethylene. Wear **30**, 349–364 (1974)
6. Muratoglu, O., Bragdon, C., O'Connor, D., Jasty, M., Harris, W., Gul, R., McGarry, F.: Unified wear model for highly crosslinked ultra-high molecular weight polyethylenes (UHMWPE). Biomaterials **20**(16), 1463–1470 (1999)
7. McKellop, H., Shen, F.W., Lu, B., Campbell, P., Salovey, R.: Development of an extremely wear-resistant ultra high molecular weight polyethylene for total hip replacements. J. Orthop. Res. **17**(2), 157–167 (1999)
8. Wang, A., Sun, D., Yau, S., Edwards, B., Sokol, M., Essner, A., Polineni, V., Stark, C., Dumbleton, J.: Orientation softening in the deformation and wear of ultra-high molecular weight polyethylene. Wear **203**, 230–241 (1997)
9. Pruitt, L.: Deformation, yielding, fracture and fatigue behavior of conventional and highly cross-linked ultra high molecular weight polyethylene. Biomaterials **26**, 905–915 (2005)
10. Tervoort, T., Visjager, J., Smith, P.: On abrasive wear of polyethylene. Macromolecules **35**, 8467–8471 (2002)
11. Lucas, A., Ambrósio, J., Otaguro, H., Costa, L., Agnelli, J.: Abrasive wear of HDPE/UHMWPE blends. Wear **270**, 576–583 (2011)
12. Jacobs, O., Mentz, N., Poeppel, A., Schulte, K.: Sliding wear performance of HD–PE reinforced by continuous UHMWPE fibers. Wear **244**, 20–28 (2000)
13. Awatani, J., Minegaki, M.: Measurements of effects of γ-ray irradiation on polyethylene by a vibration reed method. Trans. Jpn. Soc. Mech. Eng. **7**(26), 302–309 (1964)
14. Matsubara, K., Watanabe, M.: The wear properties of high density polyethylene irradiated by gamma rays. Wear **10**, 214–222 (1967)
15. Kampouris, E., Andreopoulus, A.: The effect of the gel content of crosslinked polyethylene on its physical properties. Eur. Polym. J. **25**(3), 321–324 (1989)
16. Sakoda, H., Voice, A.M., McEwen, H., Isaac, G., Hardaker, C., Wroblewski, B., Fisher, J.: A comparison of the wear and physical properties of silane cross-linked polyethylene and ultra-high molecular weight polyethylene. J Arthroplast. **16**(8), 1018–1023 (2001)
17. Atkinson, J., Cicek, R.: Silane crosslinked polyethylene for prosthetic applications: II. Creep and wear behaviour and a preliminary moulding test. Biomaterials **5**(6), 326–335 (1984)
18. Tuckart, W., Rosevear, M., Molinari, E., Gregorio, M., Failla, M.: Sliding wear behaviour of high density polyethylene cross-linked by gamma radiation. Rev. Lat. Am. Metal. Mat. **32**(2), 261–268 (2012)
19. Jones Jr, W., Hady, W., Crugnola, A.: Effect of γ irradiation on the friction and wear of ultrahigh molecular weight polyethylene. Wear **70**(1), 77–92 (1981)
20. Budinski, K.: Resistance to particle abrasion of selected plastics. Wear **203–204**, 302–309 (1997)
21. Quinn Jr, F., Mandelkern, L.: Additions and corrections: thermodynamics of crystallization in high polymers: polyethylene. Am. Chem. Soc. **81**, 6533 (1959)
22. Perez, C., Vallés, E., Failla, M.: The effect of post-irradiation annealing on the crosslinking of high density polyethylene induced by gamma-radiation. Radiat. Phys. Chem. **79**(6), 710–717 (2010)
23. Lawton, E., Powell, R., Balwit, J.: Effect of physical state during the electron irradiation of hydrocarbon polymers. Part I. The influence of physical state on reactions occurring in polyethylene during and following the irradiation. J. Polym. Sci. **32**(125), 257–275 (1958)
24. Kang, H., Saito, O., Dole, M.: The radiation chemistry of polyethylene. Temperature coefficient of cross-linking and other effects. J. Am. Chem. Soc. **89**(9), 1980–1986 (1967)
25. Dole, M.: Cross-linking and crystallinity in irradiated polyethylene. Polym. Plast. Technol. Eng. **13**(1), 41–64 (1979)
26. Henderson, P., Wallace, A.: Hardness and creep of crosslinked polyethylene. Polymer **30**, 2209–2214 (1989)
27. Baltá-Calleja, F.J.: Dependence of micro-indentation hardness on the superstructure of polyethylene. Colloid Polym. Sci. **254**, 258–266 (1976)
28. Briscoe, B., Sinha, S.: Wear of polymers. J. Eng. Tribol. **216**, 401–413 (2002)
29. Myshkin, N.K., Kovalev, A.: Tribology of polymers adhesion and friction of polymers. Tribol. Int. **38**, 910–921 (2005)
30. Bahadur, S., Stiglich, J.: The wear of high density polyethylene sliding against steel surface. Wear **68**, 85–95 (1981)
31. Sinha, K., Briscoe, B.: Polymer Tribology. Imperial College Press, London (2009)
32. Bahadur, S.: The development of transfer layers and their role in polymer tribology. Wear **245**, 92–99 (2000)
33. Briscoe, B., Pogosian, A., Tabor, D.: The friction and wear of high density polyethylene: the action of lead oxide and copper oxide fillers. Wear **27**, 19–34 (1974)
34. da Silva, C., Tanaka, D., Sinatora, A.: The effect of load and relative humidity on friction coefficient between high density polyethylene on galvanized steel—preliminary results. Wear **225–229**, 339–342 (1999)
35. Braithwaite, E.R.: Lubrication and Lubricants. Elsevier, Amsterdam (1967)
36. Lancaster, J.K.: Abrasive wear of polymers. Wear **14**, 223–239 (1969)
37. Hutchings, I.M.: Tribology, Friction and Wear of Engineering Materials. CRC Press, Boca Raton (1992)
38. Lancaster, J.K.: Relationships between the wear of polymers and their mechanical properties. Proc. Inst. Mech. Eng. **183**, 98–106 (1968)
39. Vallés, E., Failla, M.: The effect of temperature on the tensile mechanical behavior of irradiated linear polyethylene. J. Appl. Polym. Sci. **88**(8), 1925–1936 (2003)
40. Muratoglu, O., Bragdon, C., O'Connor, D., Jasty, M., Harris, W., Gul, R., McGarry, F.: Unified wear model for highly crosslinked ultra-high molecular weight polyethylenes (UHMWPE). Biomaterials **20**, 1463–1470 (1999)
41. Ting-Yung, H., Eiss Jr., N.S.: The effects of molecular weight and cooling rate on fine structure, stress–strain behavior and wear of polytetrafluoroethylene. Wear **84**, 203–215 (1983)
42. Kanaga Karuppiah, K., Bruck, A., Sundararajan, S., Wang, J., Lin, Z., Xu, Z., Li, X.: Friction and wear behavior of ultra-high molecular weight polyethylene as a function of polymer crystallinity. Acta Biomater. **4**, 1401–1410 (2008)

43. Komoto, T., Tanaka, K., Hironaka, S., Matsumoto, T., Takano, N.: Morphological study of the wear of crystalline polymers I: high density polyethylene. Wear **75**, 173–182 (1982)
44. Spalding, M., Hyun, K.: Polymer coefficients of dynamic friction as a function of temperature, pressure, and velocity for several polyethylene resins. Eng. Sci. **35**(7), 557–563 (1995)
45. Mathew, M., Novo, J., Rocha, L., Covas, J., Gomes, J.: Tribological, rheological and mechanical characterization of polymer blends for ropes and nets. Tribol. Int. **43**, 1400–1409 (2010)
46. Santner, E., Czichos, H.: Tribology of polymers. Tribol. Int. **22**, 103–109 (1989)