JOURNAL OF MATERIALS SCIENCE **39** (2004) 5625–5632

Review
Advances in the study on wear of metals by polymers

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Wear of metals by polymers is such a subject being ignored for a long time. In order to raise general concern, a review of its developments in the past decades is made, in which the emphasis is mainly put on the wear behavior and the wear mechanisms of metals by plastics and rubbers respectively. Although the essential details were revealed initially, many respects are still obscure to us, as this problem is complicated. Therefore, there is clearly a need for further study.

1. Introduction

It is well known that the wear value of a soft solid surface is usually larger than that of its hard counter face in the friction couples. However, the wear of metals by polymers, an interesting and unexpected wear phenomenon, was observed for some polymer-metal friction pairs. Unfortunately, it escaped the attention all along, even though this special wear phenomenon was discovered as early as 1960's [1, 2].

In consideration of that the polymer-metal frictional couples are used widely in a variety of machinery, moreover, the metal components of these frictional pairs are usually more expensive than the polymeric counter parts, the study in wear of metals by polymeric materials is of vital importance. Therefore, this paper is aimed to review its developments in the past decades in order to raise general concern for such a long-term ignored subject.

2. Wear of metals by plastics

2.1. Wear behavior of metals by plastics

By using a four-ball friction unit, Vinogradov *et al.* [2] found that the wear value of steel ball by plastic powders depends on its own hardness, the type of plastics and the applied load. At low load, powdered diphenyl causes the steel ball to undergo mild wear. However, the wear value of steel ball caused by polymethylmethacrylate (PMMA) is lower than that by diphenyl. As for the wear of steel ball caused by unstabilized and stabilized polypropylene (PE) respectively, their wear values are very small and decreased with the increase in the degree of branching of polypropylene. Gorokhovskii *et al.* [3] observed that the wear rate of a steel plate was increased by 116% when 5% of polyisobutylene (PIB) was added to the abrasive powder (W2B5) during the steel plate sliding against PMMA. This effect was more obvious in argon atmosphere than in air, which was however reversed when over 10% of polyisobutylene were added, and then some reduction in wear-rate of steel was observed.

Evdokimov *et al.* [4] proposed that the wear of steel by plastics varied with the type of materials, for example, the wear value of steel is increased by polyethylene and decreased by Textolite (a plastic bearing material). Zaitsev [5] has the same viewpoint according to the experimental results of wear of hard alloy (WC6-M) by polymers. High wear values were measured for the wear of hard alloy by polymers of being contained reactive groups, such as amide, ether, ester and hydroxyl group. However, no wear was observed for the hard alloy when it was rubbed against the high-density polythylene (HDPE) and polytetrafluoroethylene (PTFE) without reactive groups (Table I). From Table I, it can be also seen that the frictional coefficient, wear rate and friction temperature are related to the relative area of the friction transfer layer (FTL) in a definite mode.

2.2. Wear mechanisms of metals by plastics

During a metal ring sliding against a plastic disc in air, Vinogradov *et al.* [2] observed the mutual transfer of materials being occurred in the contact region. The plastics were moved on to the metal surface and then formed a polymer film as the thermo-oxidation degradation products of macromolecular chains of plastics could react with the steel surface, especially, with the newly-formed surface of the steel ring. When once the transfer film was peeled off, the wear of metal occurred.

Gorokhovskii *et al.* [3] considered that free radicals generated by mechanical rupture of polymer molecules were absorbed by the steel surface and caused the steel surface layer to be embrittled. Free radicals species are known to be formed when polymers molecules are fractured and their subsequent reactions are quite different in the presence or absence of oxygen [6]. In contrast,

TABLE I Friction characteristic of WC6-M alloy rubbed against polymer materials [5]

Characteristic	HDPE	PTFE	PCA	EDC	PF
Wear rate ($\times 10^{-11}$ mm^{-1})	<1	<1	6	6.6	3.1
Friction coefficient	0.14	0.098	0.19	0.22	0.16
Temperature (°C)	90–110	110–115	100–140	90–150	100–160
Relative area of FTL (%)	7–25	17–30	35–80	45–95	20–35

the wear steel surface is decreased in the presence of polypropylene radicals. Moreover, the plastic materials can in fact cause the steel either increased or reduced wear, depending on the way in which the radicals generated combine with steel [4]. Evdokimov *et al.* [4] suggested that the polymer radicals could react with positive ions, i.e., iron or carbon ions, in the steel surface and that the resulting species either diffuse into the steel or dissolve in the plastic. In the former case, as with polyethylene or polypropylene, the carbon content of the steel increased dramatically while friction and wear were reduced. However, in the latter case, as with Textolite, the steel surface was found to have an abnormally low carbon content and the wear-rate was extremely high.

As for the wear of hard alloy by polymers, Zaitsev [5] considered that it is attributed to the abrasive wear and fatigue wear of transfer (oxide) films. During the wear process, the hard alloy surface was oxidized and reacted with the macromolecular radicals of polymer. As a result, a FTL was formed on the surface of hard alloy, which depends on the chemical structure of plastics and the tribochemical reactions in friction zone. In the cases of the thermoplastic polymers having low softening temperature and containing no reactive groups, such as HDPE and PTFE, the chemical effect of wear of hard alloy by plastics is chemical interaction of WC6-Co and macroradicals formed by mechanical degradation with direct participation of oxygen in air. Thus it leads to the oxygen-containing groups being occurred in the frictional region. The wear process is on the atomic level. The organometallic compounds are formed and dissolved in the transfer films and debris. In the other cases of the plastics possessing more rigid structures and good thermo-physical properties, such as ethylene dichloride (EDC), phenol-formaldehyde resin (PF) and PCA, the chemical effect of wear of hard alloy by plastics is chemical interaction of carbonyl, ester, hydroxyl groups and also of oxygen in air with tungsten carbide and cobalt. As a result, the thin films of oxides and organometallic polymers are formed on the hard alloy surface. Zaitsev [7] and Pleskachevsky *et al.* [8] also pointed out that the degree of local oxidation of metal surface is the dominant factor for the reaction of metal itself and its oxides to polymers for steel LPB-45, LPP-hard alloy and HDPE-BK6M frictional pairs.

3. Wear of metals by rubber

3.1. Wear behavior of metals by rubber

King *et al.* [9] investigated the wear of metals by elastomers using a modified pin-on-disc apparatus. In the experiments, a metal ball slides against a disc of a clean polymer or elastomer in the presence of a clean fluid or in a dispersion of abrasive particle. The various elastomers used are listed in Table II and some thermoplastic polymers were also included for comparison purpose.

It has been found that the wear of metals by elastomers is dependent on the state of particles on the counter face (free abrasive or embedded abrasive), and the Shore hardness or elastic modulus of the elastomer. The wear rate of metals by elastomers in water without added abrasive was less than 10^{-7} mm^{-3} N^{-1} m^{-1}. When abrasive was introduced, the wear rates of metal plotted against Shore hardness of various elastomers approximates to a power relation (Fig. 1). Moreover, the rates of wear of metals increases with the increase in elastomer modulus up to about 20 MPa, but then become approximately constant value for more rigid materials (thermoplastics) (Fig. 2). However, the relations between the wear rates of metals by elastomers and elastomer resilience are reversed and just significant at around the 0.1% level (Fig. 3). In Figs 1–3, the numbers refer to the individual polymers listed in Table II.

Based on the above results, King *et al.* [9] considered that the Shore hardness or elastic modulus of elastomers is a particularly important parameter influencing the rates of wear of metals by elastomers because its value determines the depth to which an abrasive particle penetrates into the elastomer surface under load. Moreover, they proposed three factors influencing the wear-rates of metals by elastomers in an abrasive environment: (1) the total amount of abrasive being embedded in elastomer; (2) the penetration depth of the abrasive particles into the elastomer or polymer under load. This is the most important influencing factor; (3) the strength of adhesion of the embedded particles to the substrate.

AB-Malek *et al.* [10] examined the wear of alloy-steel by rubber, by using various cylindrical indenters made from alloy-steel to puncture the surface of rubber blocks repeatedly. They proposed that the dominant factors influencing the wear rate of metal by rubber include the hardness of both metal and rubber, the carbon concentration and crosslinking level of rubber, the self-lubricating ability of rubbery boundary layer and the stability of metal-oxide layer. Charrier *et al.* [11] observed that a metal needle could be worn by elastomer, if the needle penetrated the rubber repeatedly.

Zhang *et al.* [12, 13] investigated the wear of steel 45 by natural rubber (NR), nitrile rubber (NBR) and butadiene styrene rubber (SBR) respectively in three different media (air, water and NaOH solution). Based on their experimental data, three kinds of rubbers are arranged in decreasing order of wear of steel by rubber as follows: SBR > NBR > NR in air and NBR > SBR > NR in water or in NaOH respectively (Table III). The wear rate of steel by rubber is mainly dependent on the media, the Shore hardness and structure of macromolecular chain of rubber. The wear values of steel by SBR and NBR are greater than that by NR as the NR is softer than SBR and NBR. Zhang *et al.* [14, 15] also studied the wear of tool steel T10 by rubber (NBR, SBR and fluororubber) in air and in paraffin mineral oil with

TABLE II Experimental materials [9]

No.	Type		Hardness (Shore A)	Resilience (%)	Elastic modulus (MPa)
Elastomers					
1	Natural rubber		38	48	1.5
2	Epichlorhydrin (Herclar)		77	4	7.5
3	Styrene-butadiene (SBR)		73	36	6
4	Polyurethane		85	4	12.5
5	Polychloroprene (Neoprene)		68	13	4.9
6	Polyacrylate (Krynac 882 × 2)		82	4	10
7	Polysulphide (Thiokol ST)		75	28	7
8	Chlorosulphonated polyethylene (Hypalon)		84	6	12
9	Fluorocarbon (Viton B)		75	4	6.7
10	(different degrees of cure)		75	4	7
11			82	6	10
12	Silicone	(Silastic 35)	42	41	1.7
13	(different filler contents)		55	41	3
14		(Silastic 55)	56	36	3
15		(Silastic 75)	60	38	3.5
16	Nitrile (different filler contents and various ratios of acrylonitrile to butadiene)		62	7	3.8
17		(Hycar 1002)	68	16	5
18		(Krynac 802)	71	17	5.5
19		(Hycar 1041)	76	5	7
Thermoplastic					
A	Polytetrafluoroethylene				490
B	Polypropylene				1120
C	Acrylonitrile-butadiene-styrene				1400
D	Acetal				2870
E	Polyethylene (LD)				250
F	Thermoplastic polyurethane				470
G	Plasticized polyvinyl chloride				15

TABLE III Wear value of steel 45 by three kinds of rubber materials in different media (μg/h) [12]

Elastomer	Air	Water	NaOH solution
NR	4.0	43.4	24.0
NBR	32.0	99.8	53.6
SBR	48.2	60.0	24.2

or without ZDDP. They found that the wear values of steel by three different rubbers are arranged in decreasing order as follows: SBR > NBR > fluororubber in air and NBR > SBR > fluorrubber in paraffin mineral oil with and without ZDDP respectively (Table IV). However, the influence factors for the wear of steel T10 by rubbers are similar to that for the wear of steel 45 by rubbers, but the macromolecular chains of minimal oil also affect the wear of steel T10.

TABLE IV Wear value of steel T10 by three kinds of rubber materials in different media (μg/h) [14]

Elastomer	Air	Mineral oil without ZDDP	Mineral oil 7with ZDDP
Fluororubber	10.0	4.0	3.0
NBR	36.0	64.3	57.1
SBR	44.0	40.0	38.0

3.2. Wear of mechanisms of metals by rubber

King *et al.* [9] considered that in an abrasive environment, wear of steel by rubber is resulted from loosely attached material from steel surface becoming embedded in the rubber surface during sliding process. However, AB-Malek *et al.* [10] proposed that the wear of steel by rubber is resulted from the fatigue of metal oxide under high load stress during repeated punctures. Moreover, a lubricating layer of rubbery materials was observed on the metal surface. In order to clarify this phenomenon, they carried out a series of experiments by using indenters made from titanium alloy, pure titanium (99% Ti), tool steel and tungsten carbide respectively to puncture different vulcanizate rubbers. It has been found that the molecular segments of the freshly ruptured rubber were adhered to the metal surface under the action of van der Waals' secondary intermolecular forces, and then a lubricating adhesion layer of rubbery materials was formed on the indenter surface. The free radicals of segments in the lubricating layer reacted with the metal-oxide surface and produced a metal-oxide polymer complex, which is weaker than the metal-oxide surface itself and can be detached more easily from the surface. A general trend is towards increasing the wear rate with decreasing the hardness of metal because a hard metal means in fact a high and local plastic yield stress. Therefore, the stable oxide layers of metal react much less readily with polymer radicals and a relatively

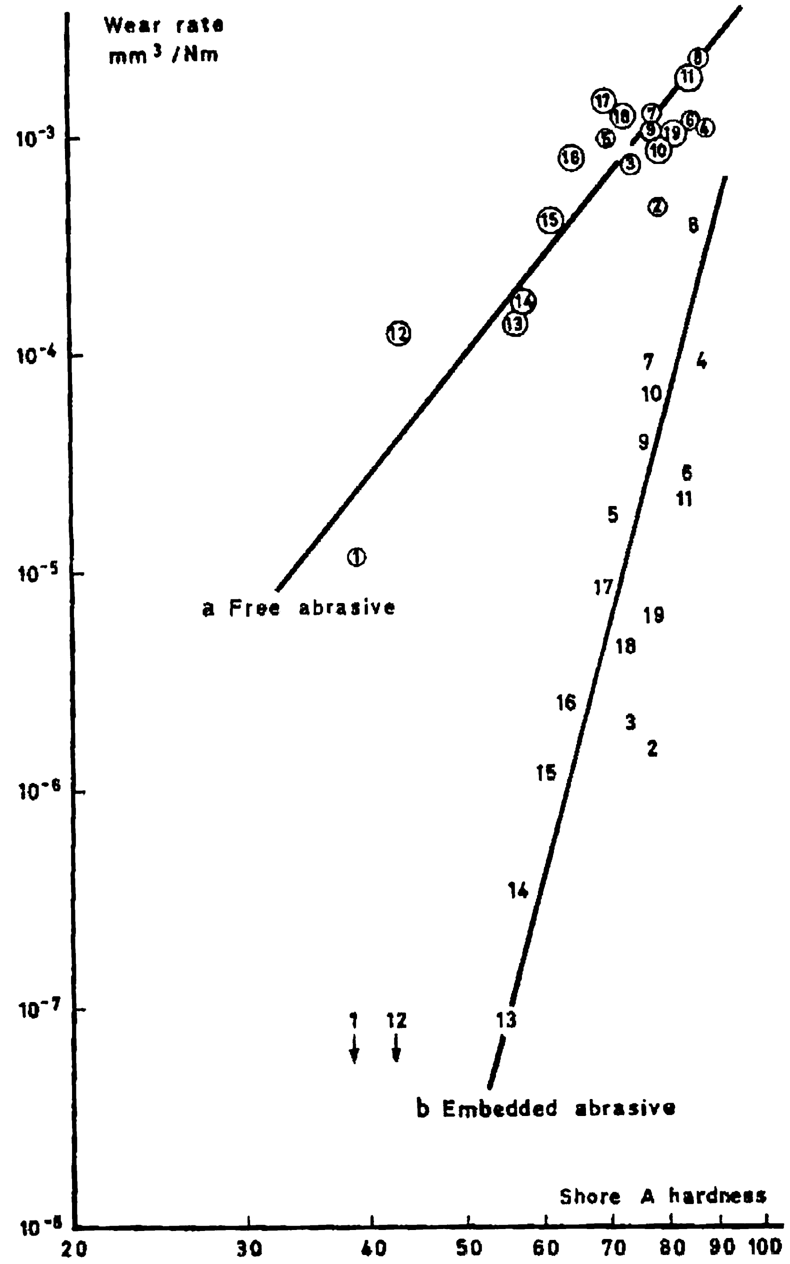

Figure 1 Variation of metal wear with elastomer hardness: curve (a), steel, free abrasive, wear rate proportional to $H^{5.2}$, correlation coefficient 0.90; curve (b), bronze, embedded abrasive, wear rate proportional to $H^{10.2}$, correlation coefficient 0.87 [9].

thick and continuous layer of rubbery material acts as a lubricant to retard wearing. But it is an anomaly in that the softest metal (titanium) shows much lower wear rates than expected for its hardness because it is noted for the stability of its oxide layer.

Charrier *et al.* [11] also found that the transfer layer of rubber on needle surface can reduce the wear of needle.

Zhang and coworkers [12–15] proposed that the wear mechanisms of steel 45 and steel T10 by rubbers respectively under the lubrication of inorganic medium or mineral oil are microcutting the steel surface by the debris and the particles of additives, in addition to the plastic deformation of metal surface. They perceived that the wear process of metal might be divided into two stages as follows. (1) The mechanochemical reactions among metal, rubber and medium result in a chemical reaction film to be formed on the metal surface; (2) The chemical reaction film of being peeled off by the microcutting action of harder particles leads to wear of metal. Therefore, for investigating the wear of metal by rubbers, attention should be paid not only to the reaction of carbon free radicals to metal but to the surfacial mechanical action as well.

Gent *et al.* [16] investigated the wear mechanism of steel by cis-polyisoprene (IR) and cis-polybutadiene (BR) respectively using a blade abrader designed by Thomas [17]. They observed that the wear of steel blade by a rubber wheel took place much more rapidly on a cis-polyisoprene surface than that on a cis-polybutadiene surface, and more seriously in inert atmosphere than in air. The long-lived radicals formed in IR appear to react readily with steel, presumably forming an iron-carbon compound, which is removed along with the rubber debris. In contrast, the more reactive radicals formed in BR appear to undergo mainly reactions within the rubber so that the steel suffers much less wear in this case. As for the peroxy radicals, the

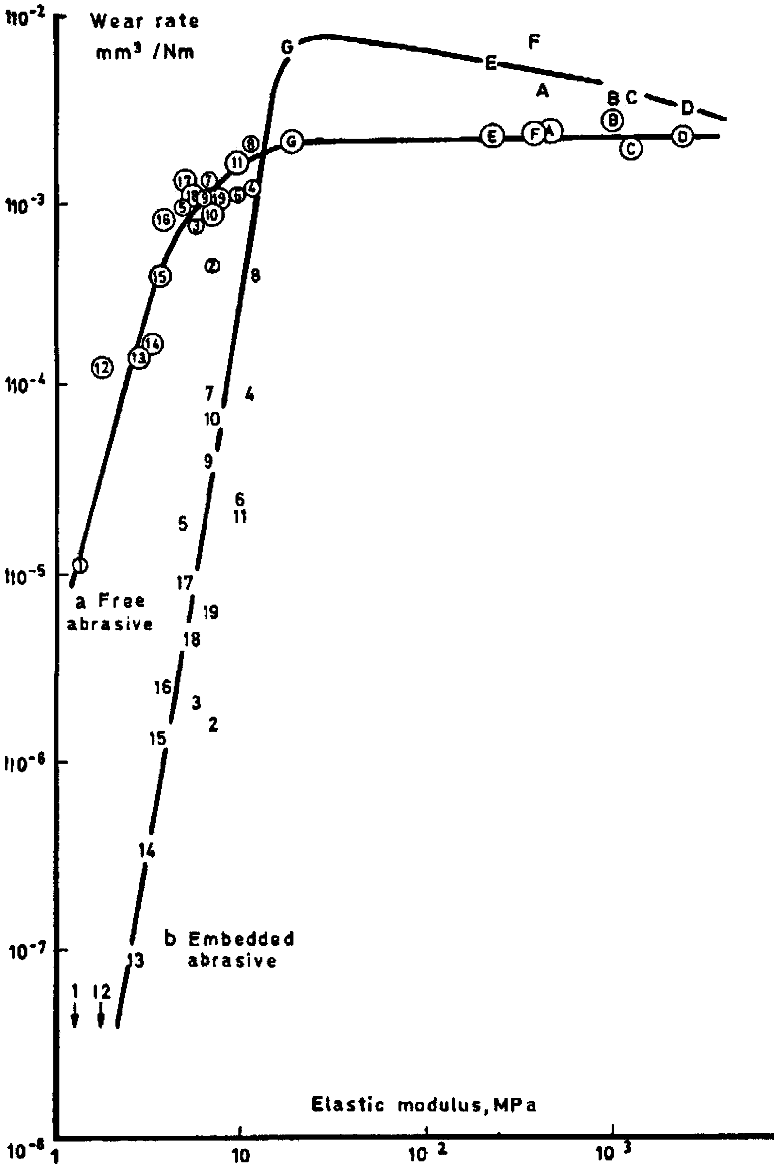

Figure 2 Variation of metal wear with elastic modulus for elastomers and polymers: curve (a), steel, free abrasive; curve (b), bronze, embedded abrasive [9].

reactivity is apparently lower for BR than for IR so that attack on steel is less for material in air.

In order to examine the process of mechanochemical reaction in detail, Gent *et al.* [18] conducted further experiments by using three kinds of metal razor blades (stainless steel, nonferrous alloy and bronze) against the surfaces of six kinds of rubbers (SBR, standard Malasian rubber (SMR), isobutene-isoprene rubber (IIR), tran-1,5-polypentenamer rubber (TPR), BR and ethylene-propylene (EPR)), respectively. It has been found that the wear rate of steel by rubber can vary by a factor of 50 or so when both the hardness of the rubber and the frictional force are kept constant (Table V). Moreover, in a nitrogen atmosphere, the wear rate of a metal scraper is generally increased by a large factor, between 5 and 50 times, depending on the rubber against which the scraper slides. It is attributed mainly to the greater stability of carbon radicals, in general, in comparison with corresponding peroxy radicals. However, an apparent exception is the rate of wear against IIR compound, it was found to be greater in air than in nitrogen, by a factor of about 3. This anomalous behavior is ascribed to enhanced stability of the peroxy radicals in IIR, compared to the carbon radicals formed by molecular rupture.

Gent and coworkers [18] pointed out that the wear rate of metal is closely related with the stability of

TABLE V Wear rate of steel razor blades [18]

Elastomer	Hardness (Shore A)	Test atmosphere	Wear rate ($\times 10^{16}$ m^3/rev)
SBR	75	Air	13
SBR	75	N_2	100
SMR	60	Air	0.75
SMR	60	N_2	40
IIR	57	Air	17
IIR	57	N_2	5.2
TPR	67	Air	0.70
TPR	67	N_2	5.2
BR	66	Air	0.30
BR	66	N_2	2.5
EPR	56	Air	0.25
EPR	56	N_2	1.3

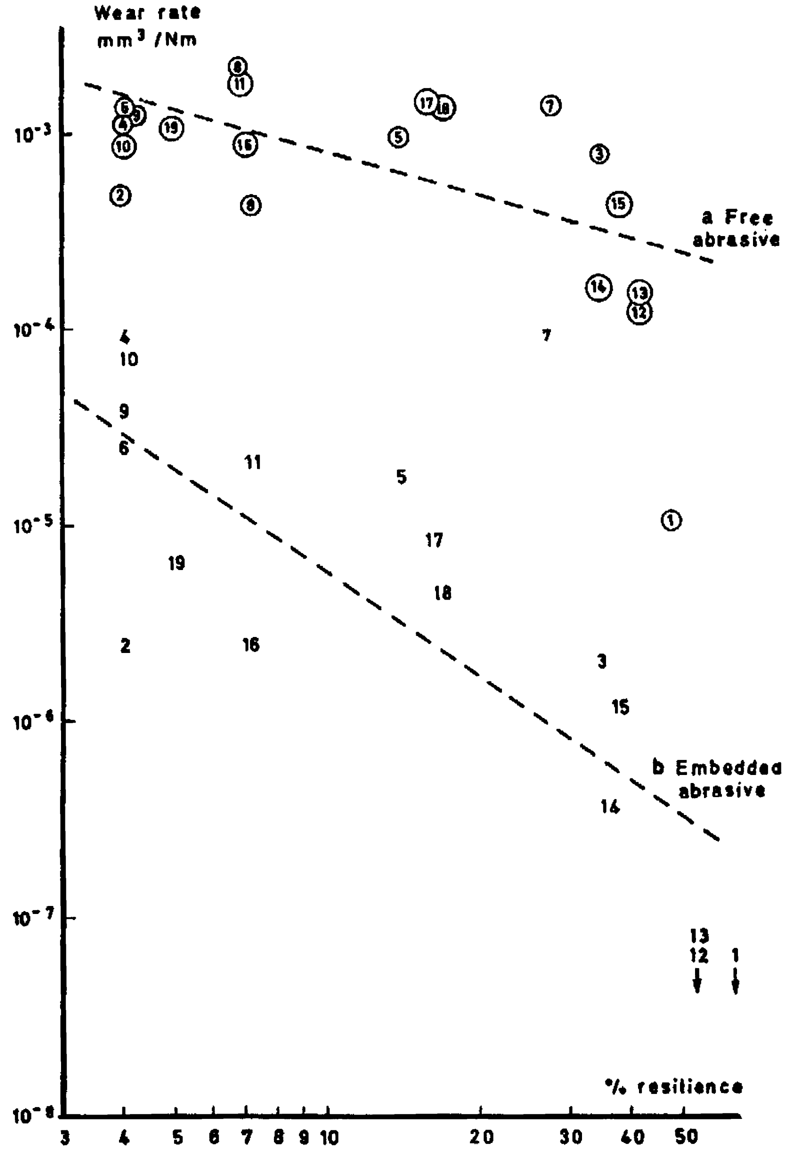

Figure 3 Variation of metal wear with elastomer resilience: curve (a), steel, free abrasive, wear rate proportional to $R^{-0.8}$, correlation coefficient −0.62; curve (b), bronze, embedded abrasive, wear rate proportional to $R^{-1.85}$, correlation coefficient −0.70 [9].

polymeric radicals, which cause metal wear by a combination of chemical reaction and detachment of metallic fragments. When the radicals are highly reactive, it is thought to take part primarily in internal polymer reactions and thus cause relatively little wear of the metal scraper. On the other hand, relatively stable polymer radicals appear to attach metals vigorously. They also found that the rate of wear of metal increased markedly as the hardness of the rubber compound increased by incorporating more carbon black, but the mechanism has not yet been identified.

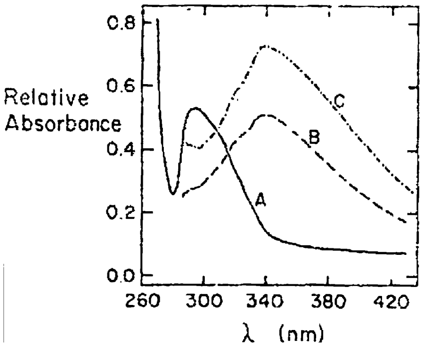

Figure 4 UV-visible spectra of styrene-butadiene rubber: (A) control sample, (B) sample sheared in air with 100 parts by weight of iron, referenced to the control, and (C) sample sheared in air with 300 parts by weight of iron, referenced to the control [19].

Some direct evidences of the reaction of macromolecular radicals with metal surfaces have been observed by Gent *et al.* [19]. In their experiments, metal powders (iron, zinc and aluminum) were incorporated into various elastomeric materials (SBR, NR, BR and EPR) and the mixtures were subjected to intense shearing in air or a nitrogen atmosphere. Fig. 4 shows the UV-visible spectra of samples of SBR, which have been subjected to intense shearing with and without iron powder being present. By using the control sample as a reference, a new absorbance is found at 340 nm for the samples that were sheared with iron powder. It is noteworthy that some iron-containing organic compounds have absorbances in this region. However, these compounds have not been identified more explicitly. As shown in Fig. 5, the absorbance at 340 nm increases

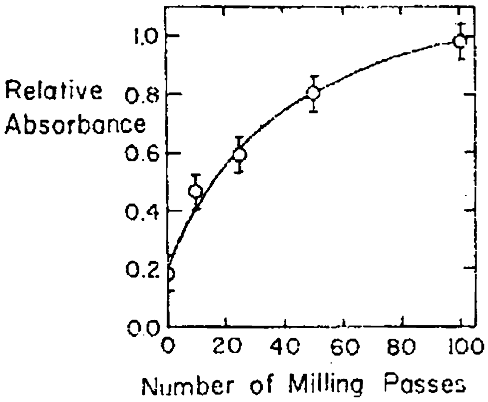

Figure 5 Relative absorbance at 340 nm versus amount of shear for styrene-butadiene rubber with iron powder (300 phr) [19].

continuously with the extent of sharing, denoted by the number of milling passes for a chemical reaction.

Gent *et al.* [19] also observed that the amount of iron or zinc taken up by the elastomer depends on the type of macroradical produced by molecular rupture, namely, relatively long-lived radicals (SBR, NR) are associated with greater metal pick-up compared with more reactive radicals (BR, EPR). These observations are in good accord with their earlier studies [16, 18].

Based on the analyses of X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectrometer (FT-IR), Zhang *et al.* [12, 13] examined the chemical mechanisms of wear of steel 45 by NR, NBR and SBR respectively. It has been found that the macroradicals produced by the rupture of molecular chains of rubber were reacted with the steel surface and generated metal-polymer complexes (Fig. 6), as a result, a chemical reaction film was formed on the steel surface.

Zhang and coworkers [12, 13] considered that the wear mechanism of metal by rubber might be regarded as chemical-mechanical self-catalytic destruction. Moreover, this mechanism are mainly under the influence of Shore hardness and the structure of molecular chain of rubber because the macroradicals are formed easily for the rubber with high Shore hardness and the stability of free radicals is depended on the structure of molecular chains. They proposed that there exists one-to-one correspondence between the stability of free radicals and the wear values of steel by rubber, thus both the stability of radicals and the wear value of steel by rubber for the above three kinds of rubber are arranged in decreasing order as SBR > NBR > NR. These results also prove that the wear of steel by rubber increases with increasing the stability of free radicals, as presented previously by Gent *et al.* [18, 19]. However, the wear of steel by NBR is most serious under the condition of a liquid medium.

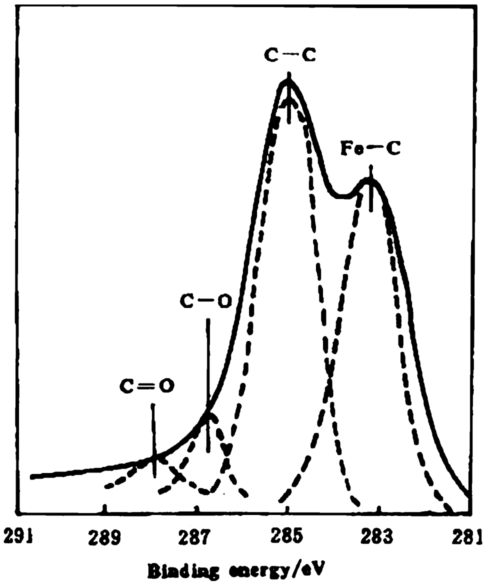

Figure 6 XPS spectrum of C element on worn surface of steel 45 [12, 13].

In addition, Zhang *et al.* [14, 15] investigated the chemical mechanisms of wear of tool steel T10 by rubbers. The graft-reaction thin films existed on the worn surface of steel were discovered firstly, which are formed by the reaction of both macroradicals and segments generated by mechanical rupture of rubber molecules during abrasion with the steel (Fig. 7). This film could improve the surface properties of steel.

By XPS analysis, Zhang and coworkers [14, 15] found that the wear value of steel by rubber is increased with decrease in the reactivity of macroradicals. When steel slides against rubber in air, both the stability of macroradicals and the wear value of steel by three types of rubber are arranged in decreasing order as SBR > NBR > fluororubber, which is in accordance with the results proposed in references [12, 13, 16, 18, 19]. By FT-IR analysis of mineral oil without ZDDP, it has been found that after metal was worn by NBR, the free radical segments of oil molecular chains were reacted to steel surface and produced Fe-polymer compound, which grafted on the steel surface. Zhang *et al.* [15] also observed that during the experiment process, mineral oil was oxidized automatically and produced some substances such as aldehyde, ketone and acid, which have catalytic oxidation action on metal. As for the mineral oil with ZDDP, the oxidation of molecular of mineral oil was inhibited so that the wear value of steel by rubber was reduced.

Recently, Wang *et al.* [20] investigated the formation of transfer film under the condition of wear of steel AISI 1020 by natural rubber. It has been found that the formation of transfer film on the worn surface of steel are involved two processes as follow. Firstly, an

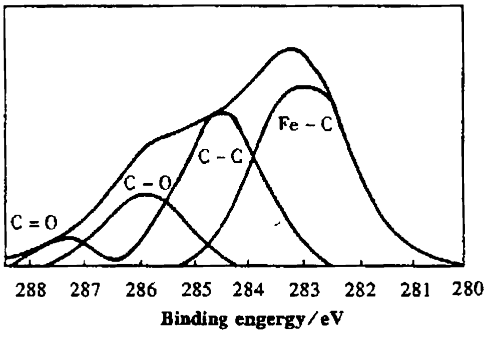

Figure 7 XPS spectrum of C element on worn surface of steel T10 [15].

adhesive layer emerged on the steel surface, which was resulted from the macromoleular chains of rubber to adhere to the steel surface by van der Waals' force. And then the iron atom and metal oxide reacted with the macroradicals of rubber in the adhesive layer and produced Fe-polymer compound. Hence, a transfer film was formed on the worn surface of steel.

4. Concluding remarks

The wear process of metals by polymers is a complex phenomenon, which involves several processes, such as physical, mechanochemical and thermal-chemical and so on. To clarify this phenomenon is of importance to the design and use of the polymer-metal friction assemblies. Unfortunately, it has received too little attention in the scientific and engineering circles in the past decades. Although marked progress is being made in this subject of recent years, there are still some key problems, which need to be addressed. It is expected that these studies would open up the prospects for developing new techniques of metal processing and surface engineering.

Acknowledgments

This work was supported by the Research Grants from the Materials and Engineering Division of the National Science Foundation of China (Grant No. 59875062).

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Received 8 June 2003
and accepted 7 April 2004