MEASUREMENT OF WEAR AND CORROSION BY SURFACE IRRADIATION WITH CHARGED PARTICLE BEAM

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Abstract

In a series of projects conducted in an industrial enviornment by the staff of the Nuffield Cyclotron Laboratory the radioactive surface layers formed by bombardment with charged particles have been used to measure wear and corrosion in metals and alloys. The radioactivity induced in a layer of approx. 50-100 μm thick can be made sufficient to provide an indication of the removal of a few microns of the material and at the same time low enough not to pose any health hazards to the personnel.

The basis of the method is measurement of the radioactivity remaining on the irradiated specimen, rather than that removed. The changes in the residual activity can be converted into the ablation thickness by calibrating the specimen, using an electro-polishing technique.

This method has been applied to the investigation of wear in a section of a pipeline made of mild steel tubing. The installation permitted a continuous on-line monitoring of wear in the pipes.

Another application of the technique was on-line monitoring of the state of wire-drawing dies made of tungsten carbide.

A corrosive attack of sea water on copper, bronze and brass was observed by monitoring ^{65}Zn formed as a result of (p,n) reaction on ^{65}Cu. The severity of corrosion and its rate could be found after only a few days of exposure.

1. The principle of the method

The surface of most metals and their alloys when irradiated with charged particles of sufficient energy becomes slightly radioactive. The exact nature of the induced radioactivity depend upon the metal and the type of bombarding particle.

The depth of the material which is activated is slightly smaller than the penetration range of the projectiles. For the beam of charged particles available in the Nuffield Cyclotron Laboratory in Birmingham, the typical activation depths in μm are given in Table 1.

Table 1

Active depths in some materials (μm)

Material	Projectile 10 MeV protons	30 MeV ^{3}He
Steel	175	200
Tungsten Carbide	120	130
Brass	175	190
Zinc	210	230
Antimony	245	315
Cobalt	160	175
Titanium	300	340
Tungsten	110	125
Copper	175	185
Platinum	100	120
Vanadium	230	260

The values in Table 1 correspond to the activating beam directed perpendicularly to the irradiated surface. The penetration of the beam can be reduced if the beam impinges at an oblique angle to the surface. In some applications one can further reduce the penetration by decreasing the projectile energy with an attenuating foil placed in its path.

In steel, proton irradiation will produce mostly ^{56}Co from ^{56}Fe. ^{56}Co has a half-life of 77 days and produces an easily distinguishable group of gamma energies up to 3.3 MeV. Being a positron emitter it can be used for autoradiography. ^{57}Co which originates from ^{57}Fe (abundance 2.2% in natural iron) has a half-life of 270 days and may therefore be useful in the investigation of processes taking place on a slower time-scale e.g. corrosion of metal objects like earthing systems, underground and underwater pipelines, cables etc. Irradiation of steel with ^{3}He produces mostly

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Table 2

Main long-lived radioisotopes formed during irradiation of steel by charged particles
(10 MeV protons, 20 Me V deuterons and 30 MeV ^{3}He)

Reaction	Abundance %	Threshold MeV	Yield* Ci/μA.hr	Half-life	Decay
$^{56}Fe(p,n)^{56}Co$	91.7	5.44	13.2	77 d	β^{+},EC
$^{57}Fe(p,n)^{57}Co$	2.2	1.65	0.006	270 d	EC
$^{54}Fe(d,n)^{55}Co$	5.8	0	1.3	18 h	decays to ^{55}Fe
$^{54}Fe(d,p)^{55}Fe$	5.8	0		2.4 y	no
$^{56}Fe(d,3n)^{55}Co$	91.7	18.4		18 h	decays to ^{55}Fe
$^{56}Fe(d,t)^{55}Fe$	91.7	5.15		2.4 y	no
$^{56}Fe(d,n)^{57}Co$	91.7	0	10	270 d	EC
$^{57}Fe(d,2n)^{57}Co$	2.2	3.9		270 d	EC
$^{56}Fe(d,2n)^{56}Co$	91.7	7.9	40	77 d	β^{+},EC
$^{57}Fe(d,3n)^{56}Co$	2.2	15.9		77 d	β^{+},EC
$^{54}Fe(d,\alpha)^{52}Mn$	5.8	0	3.3	5.7 d	β^{+},EC
$^{54}Fe(d,2p)^{54}Mn$	5.8	2.2		312 d	EC
$^{56}Fe(d,\alpha)^{54}Mn$	91.7	0	2.7	312 d	EC
$^{57}Fe(d,\alpha n)^{54}Mn$	2.2	2.2		312 d	EC
$^{56}Fe(^{3}He,2n)^{57}Ni$	91.7	6.06	860	36 h	β^{+},EC(decays to ^{57}Co)
$^{56}Fe(^{3}He,pn)^{57}Co$	91.7	1.79	36	270 h	EC
$^{56}Fe(^{3}He,p)^{58}Co$	91.7	0	18	71 d	EC,
$^{57}Fe(^{3}He,3n)^{57}Ni$	2.2	12.3	incl.in ^{56}Fe	36 h	β^{+},EC
$^{57}Fe(^{3}He,pn)^{58}Co$	2.2	0.81	incl.in ^{56}Fe	71 d	β^{+},EC
$^{57}Fe(^{3}He,p)^{56}Co$	5.8	0	1.4	77 d	β^{+},EC
$^{54}Fe(^{3}He,n)^{56}Ni$	5.8	6.2	?	6.1 d	EC

* Referred to the beginning of irradiation

^{57}Ni, ^{57}Co and ^{56}Co. The main radioisotopes formed during irradiation of steel are in Table2.The data for 20 MeV deuteron irradiation are based on results of Dmitriev et al. (1968).

Table 2 does not include activities produced by the irradiation in alloy steels due to Mn, Cr, Ni and other additives.

The irradiation of copper with protons produces mostly ^{65}Zn (245 d). The irradiation of copper with ^{3}He results in ^{67}Ga (78 h). The bombardment with deuterons yields ^{64}Cu (13 h). It is evident that both in steel and in copper processes can be studied with a characteristic time from few hours to few years.

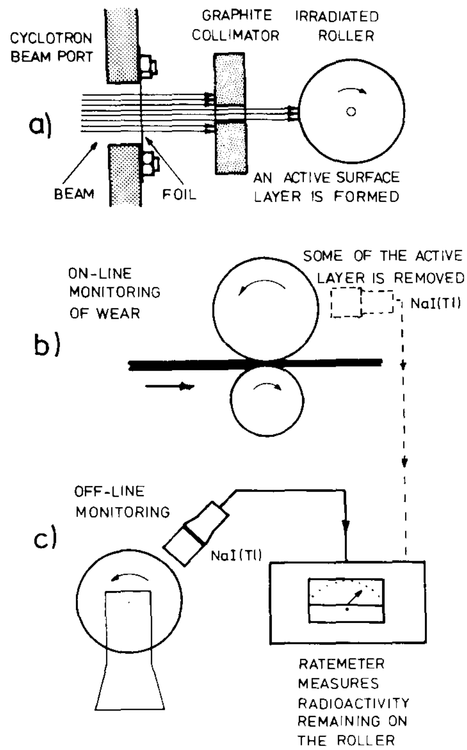

Fig. 1 The principle of radiometric method

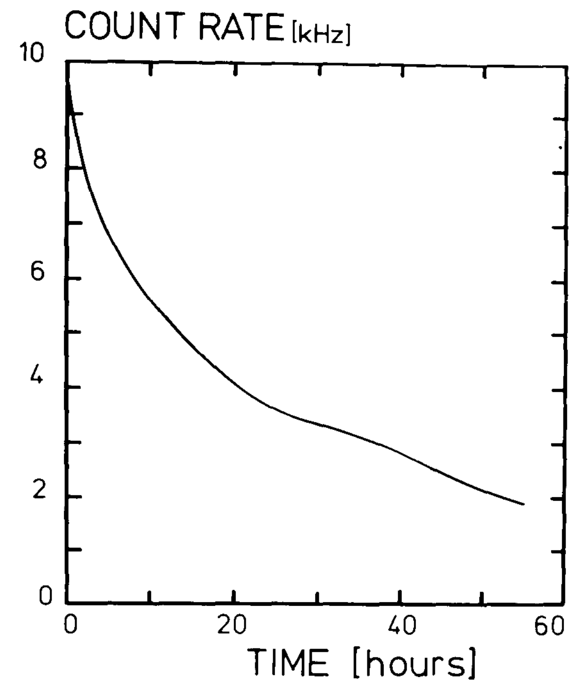

Fig. 2 Removal of radioactivity from the wall of pipeline carrying an iron ore slurry

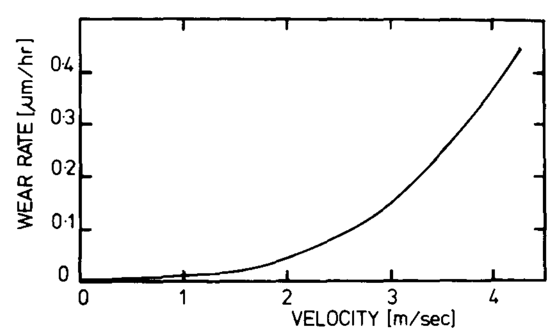

Fig. 3 The rate of wear of pipeline walls as a function of velocity of the slurry

Tin, antimony and zinc also produce useful activities.

The sample to be irradiated is placed in front of the exit port of the cyclotron (Fig. 1a). The beam spot formed on the sample can be made up to approximately 2 cm in diameter. If larger areas are to be irradiated or the uniformity of irradiation is very critical the sample can be scanned with the beam (Dealler and Ettinger, 1970). The irradiation takes place in air. The need to economize on the irradiation time may require the use of relatively high beam current that will cause local heating of the surface unless cooled by an air (or protective gas) blast. An alternative is to clamp the sample to a cooled copper block.

There is no limitation on the nature of the surface being irradiated other than the accessibility to the charged particle beam.

The irradiation is only the first part of the radiometric measurement of surface wear or corrosion. The next step is to measure how the process of wear or corrosion removes the activity from the surface (Fig. 1b).

The fact that the amount of radioactivity induced in the sample is limited only to its surface layer makes it possible to monitor the sample when actually mounted in the machinery (dies, rollers, tool bits, pipelines etc). The monitoring can be continuous or sporadic. (Fig. 1c).

To obtain quantitative results it is necessary to provide a calibration curve of the amount of radioactivity contained at various depths of the irradiated specimen. This can be done by irradiating a reference sample, then etching it electrolytically in a controlled way in suitable increments until the calibration curve can be constructed. Grinding down the sample

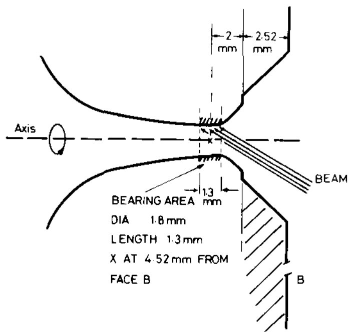

Fig. 4 The wire drawing die

surface is an alternative to etching, though less accurate.

2. Applications

The pipeline was made of mild steel and was carrying an iron ore concentrate. The cyclotron beam of ^{3}He ions was collimated with a graphite disc before impinging on the inner surface of the pipe at an angle of about 25°-30°. The sample was placed in a jig which could rotate it during irradiation. Four "hot" spots were made on each sample, separated by 90°, but a continuous ring could be made as well if necessary. After "cooling down" for a period of about 3 weeks the tubes were placed in a test rig. For the tests the rig was run with a slurry containing a 5% by volume of a coarse iron ore, having nearly 90% of iron oxides. The rig was run at a velocity of 3.3 m/sec. The radioactivity was measured with a NaI(TI) detector. The results are shown in Fig. 2 and Fig. 3. The weight loss of the radioactive section indicates a wear of 5.1 m during 50 hrs of circulation. The depth of activation was about 50 m. The useful range of wear measurement was therefore limited to about 20 m. Full details of the tests performed on pipes and of the experimental installation can be found elsewhere (Askouri, 1974).

The radiometric technique of wear measurement has been applied to wire-drawing dies, made of tungsten carbide with an addition of about 7% of cobalt. The dies were activated mounted in a rotating jig. The beam was collimated down to about 1 mm in diameter. The band of radioactivity was roughly centered on either the centre of the bearing length or at the point of minimum diameter of the die bore. The length of the "hot band" was about 1.7 mm, which was determined by means of autoradiography.

Proton irradiation of tungsten produces mainly ^{18}Re (38 d). The irradiation with ^{3}He also produces ^{184}Re as a result of (^{3}He,p) reaction on ^{182}W. Another long living component is ^{185}Os (94 d) with a very prominent 646 keV ray transition. ^{183}Re with a strong 163 keV transition is also formed during the irradiation. For the same conditions of irradiation, ^{3}He bombardment produces approximately three times greater activity and therefore ^{3}He was used in all the subsequent experiments.

The dies were run in a simple laboratory wire drawing machine. The status of the die was monitored with a NaI(TI) detector and also the diameter of the drawn wire was at intervals checked with a micrometer. Both methods did agree fairly well in new dies and in resized ones, as long as nonuniform wear did not distort the geometry.

A few samples of electrolytic copper, stock brass and naval bronze were irradiated with proton beams. The samples were then submerged in sea water at about 17°C for a period of three days. Copper and brass showed changes, but not the bronze. The change corresponds to about 2 m of the removal of surface per day. The radiometric technique offers a method to determine the efficiency of cathodic corrosion protection.

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6. Literature

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