Accelerator Based Technique of Fission Fragment Implantation for Wear Studies

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Abstract. A radiotracer technique to study wear of materials that cannot withstand a conventional surface activation by beam bombardment is presented. The thermal neutron induced fission fragments emitted from the ^{235}U layer were implanted into a sample surface in order to create a radioisotope label. Neutrons were generated by means of the ^{7}Li(p,n) ^{7}Be reaction. The proton beam from the 2.5 MeV Van de Graaf accelerator was employed. The depth distribution of the implanted fragments was determined and appeared to be ideally suited for a surface degradation study. The fragments suitable to serve as tracers were determined and their γ-spectra were measured. The degradation of the surface was derived from the activity loss. Advantages of the method are demonstrated through the application to abrasion testing of an electrostatic accelerator charge conveyer belt. The described technique may be useful for wear studies of parts made of plastics, rubber, ceramics etc.

1. Introduction

The thin layer activation technique is widely used to investigate complex physical and chemical phenomena including wear, corrosion, ion sputtering, etc. [1]. The method consists in labeling surface layers of the sample with radioisotope tracer that is followed by measurements of the γ-activity of the label. The surface degradation is derived from the activity loss. Different methods of the surface activation have been developed. All the methods can be divided into two groups: the activation of the surface by accelerated ion beams and implantation of the radioactive atoms into the surface. The first one is usually applied to metal samples. For the materials that cannot withstand irradiation with intense ion beams different alternative methods have been developed. The surface layer activation of that sort of materials can be made e.g. by isotope diffusion or by introducing recoiled radioactive particles from nuclear reactions. The possibility to employ the ^{252}Cf spontaneous fission source as an implantation device was discussed in [2]. The present paper presents a method based on the ^{235}U induced fission fragments implantation.

2. Method

A vide variety of radioactive nuclei are produced in course of the ^{235}U+n fission. Primary fragments being far from the valley of stability undergo β$^-$ decay thus producing mass chains of the same mass with gradually increasing Z. The fragments potentially suitable to serve as tracers are listed in Table I. The thermal neutron induced fission of ^{235}U is asymmetrical. Because of kinematics a lighter fragment with smaller A and Z possesses a greater fraction of the energy released in the act of fission. Because of higher energy and smaller Z the light fragments have a greater range in matter. Fission fragment ranges are rather short and so a fragment from the light group is preferable for using as a tracer. From the practical point of view an appropriate radioisotopic tracer should have a half-life of the order of tens of days. It should emit γ-rays convenient for registration, and should be produced in a significant amount.

TABLE I: CHARACTERISTICS OF SOME FRAGMENTS FOR THE ^{235}U FISSION WITH THERMAL NEUTRONS

Hal-life	Radioactive fragment	Cumulative yield	γ-ray energy, keV
1 day < $T_{1/2}$ < 8 days	^{99}Mo / ^{99m}Tc	6.11%	140.5 (89%)
	^{131}I	2.89%	364.5 (81.7%)
	^{132}Te / ^{132}I	4.3%	228.16 (88%) / 772.6 (75.6%)
	^{143}Ce	5.96%	293.3 (42.8%)
8 days < $T_{1/2}$ < 12 days	^{140}Ba/^{140}La	6.21%	537.3 (24.4%) / 328.8 (20.3%), 487.0 (45.5%), 815.8 (23.3%)
	^{147}Nd	2.25%	531.0 (13.1%)
$T_{1/2} \approx 30$ days	^{141}Ce	5.85%	145.4 (48.2%)
	^{103}Ru	3.03%	497.1 (90.9%)
$T_{1/2} \approx 60$ days	^{95}Zr / ^{95}Nb	6.50%	724.2 (44.17%), 756.7 (54%) / 765.8 (100%)
$T_{1/2} \approx 300$ days	^{144}Ce	5.50%	133.2 (11.09%)

Given these requirements the mass chain
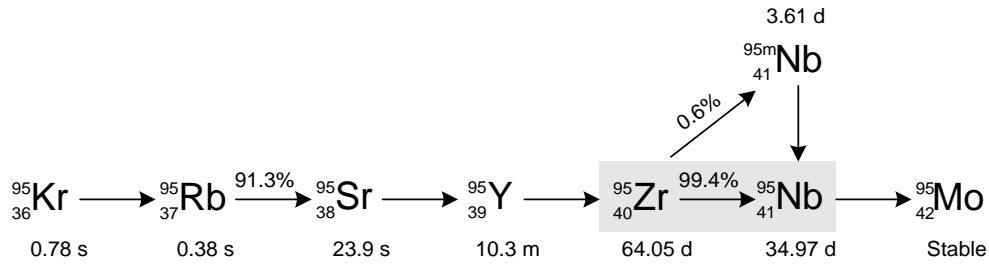
was found to produce a pair of the most suitable nuclei ^{95}Zr (half-life $T_{1/2}$=64.05 d, cumulative yield of 6.5%) and ^{95}Nb ($T_{1/2}$=34.97 d), the latter being a product of the beta decay of the former. The beta-decay of ^{95}Zr is accompanied by emitting γ-rays of 724.2 keV (44.17%) and 756.7 keV (54%) and for the decay of ^{95}Nb the γ-ray energy is 765.8 keV (100%). Since ^{95}Zr is a parent nucleus for ^{95}Nb the concentration N_{Nb} of the ^{95}Nb nucleus increases as ^{95}Zr decays according to the following equation

$$N_{Nb}(t) = N_{Nb}^0 e^{-\lambda_{Nb}t} + \frac{\lambda_{Zr}N_{Zr}^0}{\lambda_{Nb} - \lambda_{Zr}}(e^{-\lambda_{Zr}t} - e^{-\lambda_{Nb}t}), \tag{1}$$

where N^0_{Zr} and N^0_{Nb} are the initial concentrations of ^{95}Zr and ^{95}Nb, respectively and λ=ln2/$T_{1/2}$. As a result a total activity of the three γ-rays remains to be at a measurable level for a long time (*FIG. 1*).
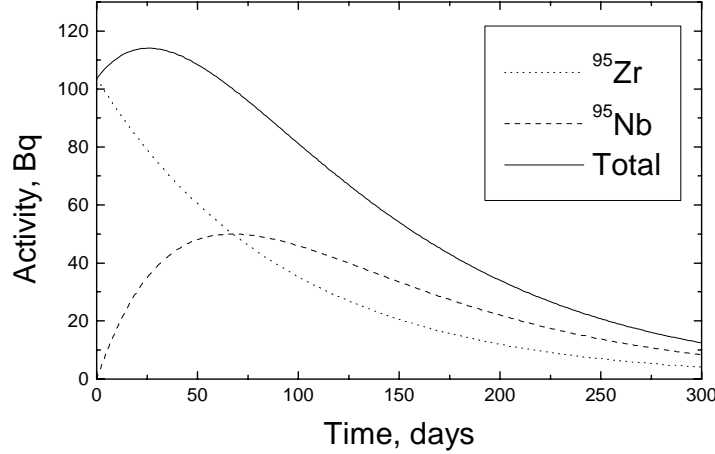
FIG. 1. Dependence of the activity of the ^{95}Zr and ^{95}Nb isotopes on time calculated from Eq. (1) for the experimental conditions of the present work.

This makes it possible to postpone the surface degradation study until most of the other implanted fragments completely decay that provides favorable background conditions for the γ-spectrometry.

If plural scattering of fragments in the uranium layer is neglected then the number of fragments dN emitted in the direction defined by a polar angle θ within a solid angle $d\Omega = 2\pi\sin\theta d\theta$ is constant:

$$dN = const \cdot \sin\theta d\theta . \tag{2}$$

Suppose the uranium layer is located in a close proximity to the irradiated surface. In simplified assumptions of a single kinetic energy specific for each of species and absence of straggling all the fragments of the same sort will have a unique range R and will stop at depth

$$x = R \cdot \cos\theta , \tag{3}$$

where x-axis is oriented along the normal to the surface. It follows from Eqs. (2)-(3) that the depth distribution of the implanted nuclei will then be uniform:

$$\frac{dN}{dx} = \frac{const}{R} . \tag{4}$$

In order to obtain a more realistic depth profile it was simulated by a Monte Carlo method. Lavsan (polyethylene terephthalate) as irradiated material was taken since the depth distribution of implants was experimentally determined in the present work using a stack of Lavsan films (see *FIG. 2* for the experimental layout). The simulation was made for ^{95}Sr because it has the largest individual yield of 4.5% in the 95 amu mass chain. A gap in 1 mm of air was assumed between the uranium layer and the irradiated surface to reproduce real experimental conditions of the present work. The first two moments of the energy distribution for this fragment were taken to be equal to <E>=97.8 MeV and σ_E=5.6 MeV respectively. Look-up tables of ranges and straggling for ^{95}Sr in the respective substances were calculated using SRIM program [3]. The results of the simulation are presented in *FIG. 3*. As is seen from the figure there is a relatively wide flat region in the implanted fragment depth profile that ideally suits for the surface degradation studies.
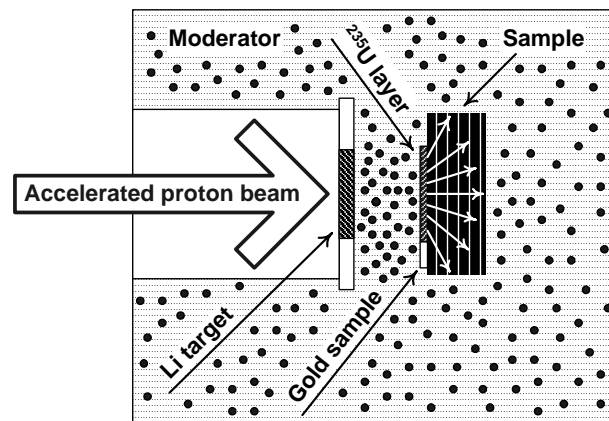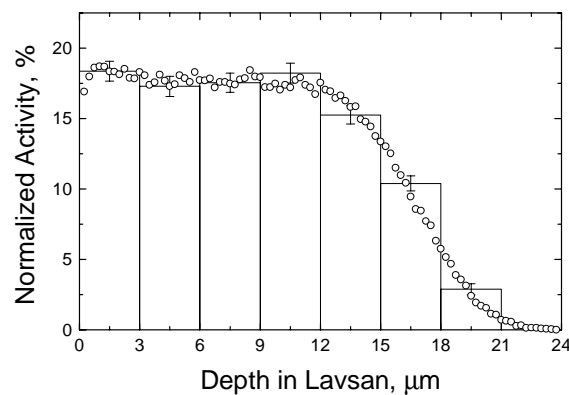
FIG. 2. Experimental layout.

FIG. 3. Measured (bar graph) and simulated (dots) depth distribution of the ^{95}Zr/^{95}Nb activity in Lavsan.

3. Experimental

A proton beam from the IPPE 2.5 MeV Van de Graaff accelerator was employed to generate neutrons by means of the ^{7}Li(p,n)^{7}Be reaction. The accelerator operated at its maximum parameters achievable at the moment: the proton energy was 2.35 MeV just above a resonance in the ^{7}Li(p,n)^{7}Be reaction and the beam current was about 7 μA. A target was made of metallic lithium of natural abundance. To avoid overheating it was cooled by flowing water. The target assembly was located in the center of a polyethylene cube with a side of 30 cm which served as a moderator of neutrons. The ^{235}U$_3$O$_8$ layer deposited on an aluminum backing was 2 mg/cm^2 thick and 30 mm in diameter.

A stack of 3μm Lavsan films was irradiated during 20 hours in order to determine the implanted fragments depth profile. The neutron flux during the irradiation was monitored by boron counter. In order to measure the fluence of neutrons a tiny gold reference specimen with known mass was placed close to the uranium layer. The absolute activity of the reference specimen was then evaluated by means of a γ-spectrometer with calibrated efficiency. The cross section for gold activation by thermal neutrons was assumed to be 98.7 barns [4]. The fluence thus determined was $2.3 \cdot 10^{12}$ neutrons/cm^2. According to calculations the total number of implanted fragments was $3.2 \cdot 10^{10}$.
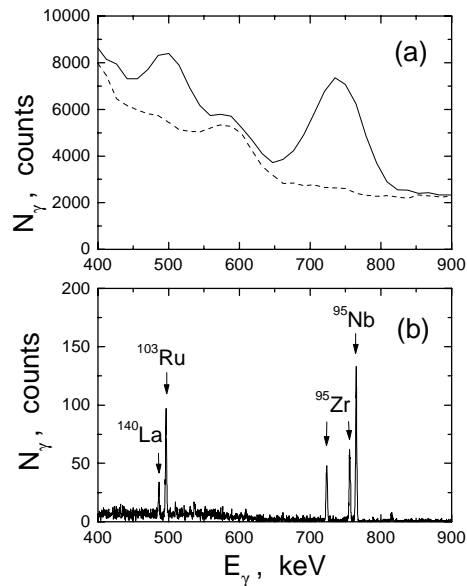
FIG. 4. Range of interest of the gamma-ray spectrum from the fragments implanted in a stack of Lavsan films obtained with NaI (a) and Ge(Li) (b) detectors on the 60th day after the implantation. The measurement time was 15 min in both cases. A dash line shows the environmental background for the spectrum measured with NaI.

Gamma-ray spectra measured from the irradiated stack with $\varnothing160\times100$ NaI and 60 cm^3 Ge(Li) detectors 60 days later the end of the implantation are shown in *FIG. 4*. As is seen from the figure the γ-rays specific for the ^{95}Zr and ^{95}Nb isotopes may be easily measured by both types of the detectors. The radiation produced by ^{103}Ru and ^{140}La isotopes used as tracers in [2] is fairly well measurable with the Ge(Li) detector but not with NaI. Comparison of the measured activity of each of the Lavsan film with results of the fragments depth profile simulation shows good agreement (see *FIG. 3*).

4. Application

The method was applied to abrasion testing of the electrostatic accelerator charge conveyer belt. There are some advantages in depositing electric charge on the electrostatic accelerator conveyer belt by brush contacts rather than by means of non-contact methods. However the attrition caused by the sliding contacts can create a problem if the belt surface material is of insufficient wear resistance. Even a microscopic deterioration of the belt surface may influence its dielectric properties and small amounts of grime gathered in surface imperfections may interfere the process of electrification. Besides dust deposition on the accelerator high voltage structure reduces its dielectric strength. The aim of the present work was to select among different belt samples the most resistant one. The samples under investigation were made of the same basic materials with some modification of the composition and technology.

The implementation of the study resembled the ordinary thin layer activation method [1] in all respects save one: the activation was made by fission fragments implantation instead of irradiation of the sample surface with an accelerated ion beam. Fission fragments were implanted into the rubber surface of the belt samples of different types and the samples were then tested at a test bench in the close to real conditions. The absence of the induced

radioactivity from the belt material irradiated by neutrons themselves was checked in a separate experiment. In 6 hours after the end of the irradiation no γ-rays exceeding environmental background was found in the measured spectrum. The implantation was made in the same manner as described above. The only difference was that for safety reasons the uranium layer was hermetically sealed in a container, the fission fragments being emitted through a window made of the 3 μm Lavsan film.

Two different types of testing have been performed. First the absolute rate of the wear was studied. The abrasion testing of the samples started in two month after the implantation and the net time of testing was 300 hours. Such a significant delay with the start of testing was caused by organizational problems. Cooling time that was needed to allow the short lived fragments to disintegrate under a level which does not influence the measurements of the selected γ-rays was about two weeks. The total activity of the samples dropped below the level that can be dangerous for the health of the personal in a day after the end of irradiation. Gamma ray spectra were measured with a Ge(Li) detector. A typical result for the residual activity as a function of time is shown in *FIG. 5*. The residual activity was converted into the thickness of the removed material using a calibration curve. The calibration curve was derived from the data presented in *FIG. 3* with a correction for the difference of the fragments ranges in Lavsan and rubber. A linear dependence of the wear on time was assumed and for the case shown in *FIG. 5* the wear rate was found to be 0.019±0.003 μm/h.

It was assumed in the present work that the implantation of the fission fragments did not affect resistibility of the sample surface to abrasion. As a matter of fact even a single energetic heavy ion creates a latent track in the matter that in principle may influence properties of the surface. Consequently some additional study is needed in any case of the method application in order to check whether the implantation dose is low enough for the mechanical properties of the surface to remain unaffected. Unfortunately the information on the influence of radiation effects in organic materials on their mechanical properties is restricted mainly by gamma-ray and electron irradiation.
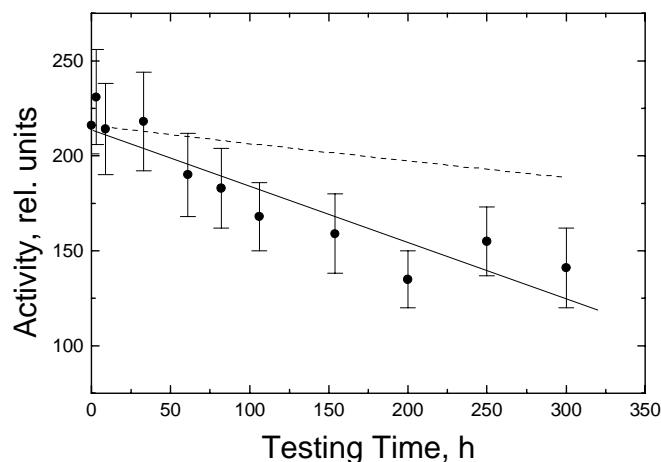

FIG. 5. Residual activity of the sample under investigation as a function of a net time of the abrasion testing. A solid line was drawn over measured points by a least-squares method. A dash line represents the activity reduction due to a natural decay of the implanted tracer.

In the other type of study only relative wear resistance of the samples was investigated. If the implantation takes hours rather than weeks (note that it lasted 46 days in [2]) radioisotopes with relatively short half-life and consequently higher activity can be used provided that the task permits short term testing. This was the case in the present study. The mass chain with the beta decay rate controlled by isotope ^{99}Mo ($T_{1/2}$=66.02 h, cumulative yield of 6.11%) produced during the labeling process was selected in order to optimize measurement parameters. The corresponding γ-ray energy of 140.5 keV (parent nucleus ^{99m}Tc) is low enough so as to be registered with high efficiency. At the same time the γ-ray penetrability is sufficient for the self-absorption in the sample to be neglected. A typical γ-spectrum for a single film measured in a day after the end of the irradiation is shown in *FIG. 6*.

Strips cut of the belt of different types were irradiated with fission fragments during 20 hours in the same geometry as described above. After the irradiation, the strips were mounted around a roller rotated by an electric motor. The roller diameter was 9 cm and the rotation speed was 1200 rpm. Sliding contacts made of a nickel foil 0.1 mm thick were pressed to the strips with an enhanced strength (as compared with used in real conditions) in order to accelerate abrasion. So far as the study was comparative only relative rate of the wear for the investigated samples was important. The abrasion testing started a day later the irradiation. The γ-ray spectra from the samples were measured in regular intervals during the abrasion and the wear kinetics as a function of time was derived from the remaining radioactivity by the help of a calibration curve. A correction for the decay of the implanted nuclei was made while the conversion of the activity change into the thickness of the removed material layer.

Typical results obtained in the study are shown in *FIG. 7* where the wear kinetics is compared for two selected samples. It was assumed that after initial period of the wear which was arbitrary taken to be equal to 2 hours the dependence of the material loss on time should be linear. The corresponding straight lines drawn by the least squares method have slope of 1.92±0.08 μm/h and 2.84±0.23 μm/h respectively. In a similar way the difference in wear resistance for all the investigated samples was determined.
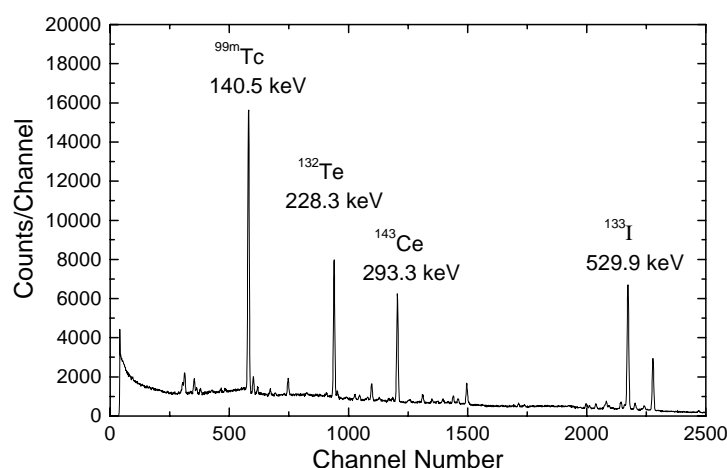
Fig. 6. Low energy part of gamma-ray spectrum showing the most intensive radiation from fission fragments implanted in a stack of Lavsan films in a day after the implantation.
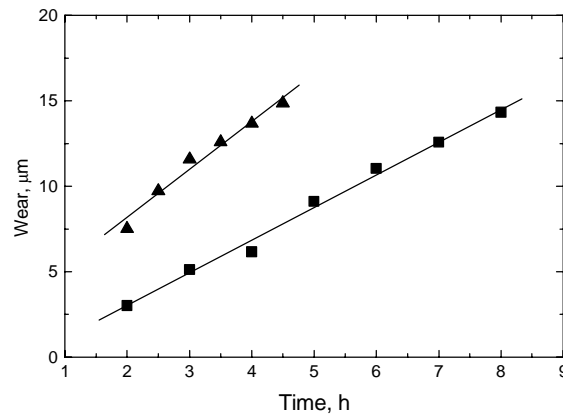
Fig. 4. Comparison of wear kinetics for a pair of the investigated samples.

5. Conclusion

The induced fission fragments implantation proved to be a convenient tool for introducing thin radioactive layers in the surface of the samples which cannot withstand activation by ion beam bombardment. A vast variety of radioactive nuclei produced in fission make it easy to find a tracer with suitable half-life and γ-ray energy. The technique is based on widely available small accelerators and is rather simple in materialization. The radioactivity introduced into the samples under investigation is extremely small and so the wear testing can be made with no danger to the environment and staff.

References

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