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^{7}Be radioactive beam production at CIRCE and its utilization in basic and applied physics

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Abstract

A pure ^{7}Be beam with an energy $E = 1–8$ MeV is available for nuclear and applied physics at the 3 MV Pelletron tandem accelerator CIRCE in Caserta. The beam is produced using an offline technique. Typical analyzed beam intensities are about 2 ppA, using cathodes with an activity of the order of 200 MBq. The ^{7}Be implantation has been used for both fundamental nuclear physics and applied physics. In particular, different metals have been implanted with ^{7}Be in order to study the influence of the chemical composition and of the number of quasi-free electrons of the host material on the ^{7}Be half-life. In the field of applied physics, the ^{7}Be implantation turns out to be very interesting for wear measurement. In fact, in this case ^{7}Be is used as a depth-sensitive tracer. The continuous detection of the sample activity during the wear allows a high sensitivity measurement of wearing speed. The ^{7}Be beam production at CIRCE, the implantation procedure and the results obtained from the ^{7}Be half-life measurements and the wear characterization of implanted steel samples are described.

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Keywords: Radioactive ion beams; Ion implantation; Wear monitoring

1. Introduction

Radioactive ion beams are acknowledged to represent one of the major challenges for nuclear physicists in future years. Large facilities are required in view of the sophisticated separation and transport systems necessary to achieve reasonably high beam intensities for short lived nuclides, which require online production. In case of medium or long lived ones, offline production is possible and this offers the opportunity to produce radioactive ion beams at small facilities. A pure ^{7}Be ($T_{1/2} = 53$ days) beam with an energy of 1–8 MeV is available at the 3 MV Pelletron tandem accelerator CIRCE (Center for Isotopic Research on the Cultural and Environmental heritage) in Caserta (Italy). The ^{7}Be implantation is delivered for different purposes. In the present work, after a short description of the technique, we report the applications of the ^{7}Be implantation in both basic and applied nuclear physics.

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2. Methods

The details of the procedure followed to produce the ^{7}Be beam are described elsewhere [1]. Briefly, the ^{7}Be nuclides are formed via the ^{7}Li(p,n)^{7}Be reaction bombarding metallic Li targets with a 11.4 MeV proton beam of 20 μA intensity, delivered by the ATOMKI cyclotron in Debrecen. Final activities are typically about 10 GBq. The irradiated Li targets are processed at the Isotopic Laboratory of the Ruhr-Universität Bochum, where the ^{7}Be nuclides are extracted from the ^{7}Li bulk following the radiochemical procedure described in [1]. The preparation takes place in a hood shielded with 10 cm of lead to protect the operator. The overall absorbed dose during each preparation, mainly concentrated in the hands and the head, is lower than 3 mSv on the hands and about 300 mSv at the rest of the body. This dose is usually shared between 2 and 3 operators. Up to 95% of the activity is collected in a liquid solution, while ^{7}Li is depleted by about six orders of magnitude. Thus the solution which is dropped in copper cathodes still contains about equal amounts of ^{7}Li and ^{7}Be.

The activities of the ^{7}Be cathodes range from 100 to 200 MBq. This amount of activity, together with the low radio-toxicity of ^{7}Be, due to its decay mode, makes it easy to fulfil the radioprotection requirements and obtain the necessary permissions.

The ^{7}Be cathodes are finally installed into a dedicated multi-sample Cs-sputtering source (model SNICS, manufacturer National Electrostatics Corporation) at the CIRCE laboratory in Caserta (Italy). Fig. 1 shows the layout of the experimental setup. The injection system, which consists of an electrostatic analyser and a dipole magnet, selects mass 23, comprising $(^7\mathrm{Be}^{16}\mathrm{O})^-$ molecules together with the Li contamination $(^7\mathrm{Li}^{16}\mathrm{O})^-$. The interaction with the gas stripper in the terminal of the accelerator breaks the molecular ions and produces a positive ion beam in multiple charge states. A post-stripper carbon foil is installed before the analysing magnet to bring ^{7}Be ions to the 4^+ charge state, which is selected with the analysing magnet in order to suppress the accompanying ^{7}Li beam. Finally the beam is analyzed and focussed by a fractioned 90° electrostatic analyser and transported to an implantation setup. The overall efficiency depends on the charge state probability, which in turn depends on the energy. For instance, the overall efficiency for a 7 MeV beam is about 9×10^{-4}. Typical current intensities are few ppA.

The ^{7}Be beam is mainly used for implantation in various materials. The implanted dose is monitored by observing the 478 keV γ-rays emitted from the sample by means of a HPGe detector. Usually the implanted ^{7}Be activities are smaller than 20 kBq.

3. Application 1: half-life in metallic environments

One of the applications of the ^{7}Be implantation has been the study of the dependence of the electron capture (EC) decay probability in different metallic environments. Assuming that the EC decay probability may be proportional to the electron density at the nucleus, several experimental efforts have been done in order to measure the variation of the ^{7}Be decay rate as a function of the environment (pressure, different chemical environments, temperature, metallic structure). Although the half-life changes are in most cases lower than 0.2% some authors found larger variations ([2,3] and references therein) in the order of few percent. Recently, the large amount of experimental data obtained in the capture cross section of light ions at low energies (d(d,p)t [4], ^{7}Li(p,α)α,^{6}Li(p,α)^{3}He [5]) in metals suggested that the quasi-free electrons in a metallic host material may modify the electron density around the

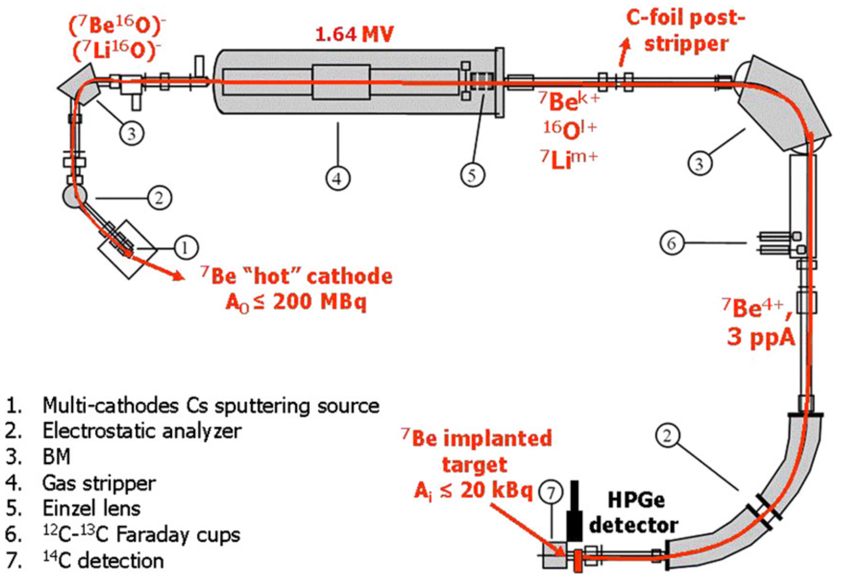

Fig. 1. Layout of the CIRCE accelerator in Caserta. The accelerator parameters are set for a 7 MeV ^{7}Be implantation. The HPGe detector measures the increasing 478 keV γ-rate of the implanted target.

implanted nuclei. In fact, a clear correlation between the measured enhancement of the electron screening effect and the number of the quasi-free electrons in the target host material was observed. In this framework, one expects that the electron density around the nuclei should change in different metallic environments and nuclei, such as ^{7}Be, which decay via electron capture are good probes for testing the electron density variations.

A crucial aspect is to incorporate ^{7}Be nuclei in a metallic environment deeper than the superficial oxidized layer and to avoid damages of its structure. This can be achieved implanting ^{7}Be at energies of a few MeV.

We implanted the ^{7}Be beam in Pd, Zr, Ta and W samples at 3–4 μm depth. The ^{7}Be half-life has been measured independently in two laboratories (Naples and Debrecen) in order to minimize systematic errors. We do not find significant variations between the samples within the experimental precision of 0.3% [6] and the absolute values are in agreement with the adopted ^{7}Be half-life.

A second implantation was done in Pd and In samples. In this case, we measured the variation of the ^{7}Be decay rate between room temperature and 12 K, achieved installing the sample at the cold head of a cryopump. We found [7] a significant variation $(-0.9 \pm 0.2)\%$ in the Pd sample and $(-0.7 \pm 0.2)\%$ in In. The minus sign means that at low temperature the ^{7}Be half-life is longer than at room temperature. We measured also a ^{7}BeO/^{7}LiO sample, where the chemical environment of the ^{7}Be is insulator-like. This sample did not show any change within the errors: $(+0.2 \pm 0.3)\%$.

A similar effect has been found also for ^{22}Na implanted in Pd $(+1.2 \pm 0.2\%)$ [8] and ^{210}Po $(+6.3 \pm 1.4\%)$ [9], i.e. for a β^+ and a α decay, respectively. Although these data are in a good qualitative agreement with the model developed to explain the observed electron screening in metals in terms of electron density, they show significant deviations from its predictions, thus suggesting that either some experimental or theoretical aspects deserve further investigation.

4. Application 2: wear measurements in material science

Wear studies with radioisotopes are a standard procedure, which is routinely used at many laboratories. γ-emissions from the radioisotopes allow online continuous wear measurements. The loss of material due to wear is proportional to the observed variation of the radioactivity previously incorporated in the sample, once its depth distribution is known.

Depth-sensitive tracers are mainly produced with the activation techniques. However, the utilization of a pure ^{7}Be beam as tracer offers a wider range of applicability and a higher sensitivity:

(1) The ^{7}Be implantation can be done in whatever material. The depth distribution of the ions is well controlled, because it depends only on the beam energy and the stopping power. Instead, the activation technique and the depth distribution of the radioactive nuclei are strongly material-dependent, following the activation cross section and the distribution of the elements into the sample.
(2) The implantation damage induced by the beam is negligible, because of the very low ^{7}Be activities needed (tens of kBq). In the activation method, due to the low activation cross sections, the required high intensity beams may modify the mechanical properties of the material.
(3) In the activation method, in most cases, it is not possible to activate selectively only one isotope and/or element.

Controlled depth profiles can be achieved varying the beam energy from the accelerator or using absorbers, as described in [10]. The maximum range of ^{7}Be ions with 8 MeV energy is lower than 10 μm, depending on the material and a submicrometric sensitivity can be achieved.

The ^{7}Be implantation technique for high sensitive wear measurements has been already applied for the wear study of polymeric and medical materials using indirect ^{7}Be implantation. In these cases, ^{7}Be can be implanted by nuclear reactions, taking place in a foil placed in front of the sample (for instance, ^{12}C(^{3}He,2α)^{7}Be [11] or ^{7}Li(p,n)^{7}Be [12]). The depth distribution of the implanted samples follows the kinetic energy of the recoil ions. The advantages of the direct implantation proposed in this work are (1) the possibility to implant higher doses (up to several kBq) increasing the sensitivity of the method and (2) the higher control of the depth distribution – directly related to the beam energy – allows to optimize the ^{7}Be depth profile following the different needs of the wear measurement.

In the framework of a joint research project between INFN, Mechanical Engineering Department (DIME) of the University Federico II of Naples and Colmegna Sud, a company specialized in thermal surface treatments, the ^{7}Be implantation technique is used for the development of new superficial hardening procedures in steel, based on the carbon diffusion.

The wear measurements are made with a pin-on-disk machine, where the ^{7}Be-implanted pin with a load on the top is blocked in a fixed position in contact with a rotating disk (Fig. 2). The disk and the pin are made up of similar treated steel material. The HPGe detector is placed in close geometry to the sample in order to increase the γ-detection efficiency. During the wear of the pin, we periodically collect the produced dust cleaning the disk. The γ-rate of the material removed during the test is also measured.

As a test of this technique and to compare it with other techniques, we implanted ^{7}Be in the steel samples at a fixed energy. In this case, the activity of the sample should decrease when the wear depth reaches the range of the ^{7}Be ions in the matter. We simultaneously measure the wear with different methods (1) the loss of weight of the pin (2) the pictures of a reference scratch on the pin surface and (3) the measurement of the removed and the residual

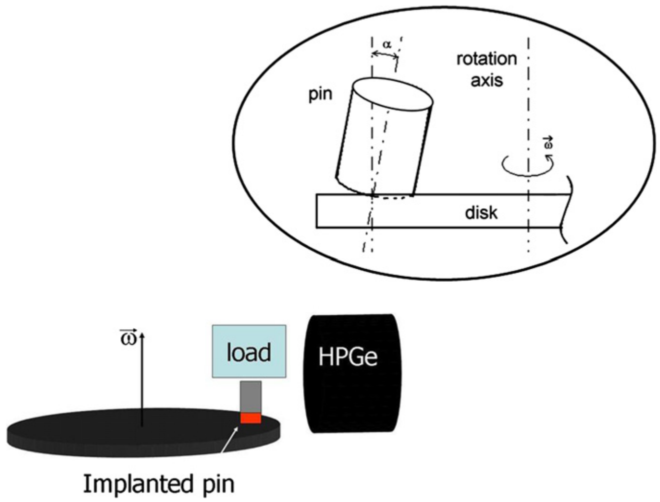

Fig. 2. Scheme of the wear measurement of a steel pin, using the pin-on-disk standard machine and the HPGe detector for the γ measurement. Angle α indicates a misalignment that causes the discrepancy between the γ-measurements and the weight shown in Fig. 3.

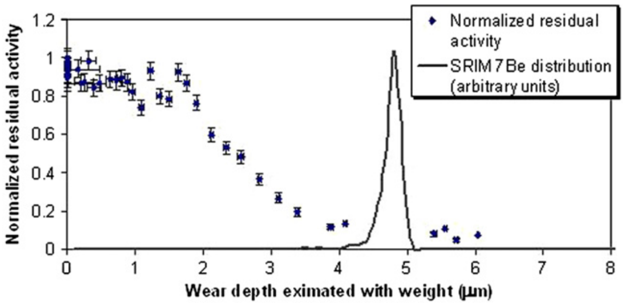

Fig. 3. Normalized residual activity of a treated steel sample as a function of the wear depth estimated with the loose of weight of the pin. The SRIM simulation of the ^{7}Be depth distribution is also shown.

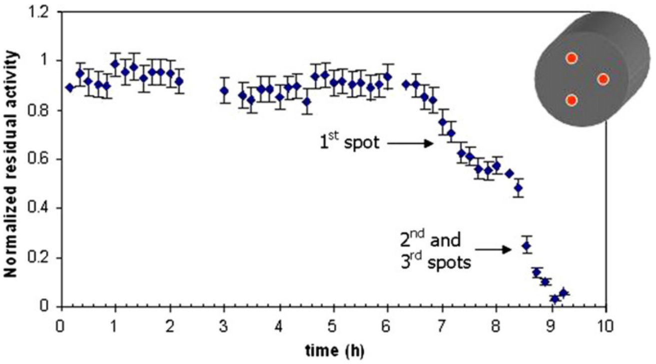

Fig. 4. Normalized residual activity of a treated steel sample as a function of time. The implantation is made at the same energy (5 μm range) on three spots. The pin is slanted in the direction of the first spot, which is seen to wear off more slowly than the other two.

activity. Fig. 3 shows the resulting behaviour of residual activity as a function of the wear depth, estimated by periodically weighing the pin during the test. Even though the qualitative trend is as expected, the residual activity decrease takes place at a smaller depth than calculated by the range of the ^{7}Be ions in steel (indicated by the energy loss peak in Fig. 3). This effect was found to be due to the inclination of the wearing surface with respect to the disk. In order to identify the plane of wear on the pin, we implanted three spots on a circle evenly spaced at 120°. Fig. 4 shows the normalized residual activity of a pin implanted on three spots as a function of time. It is easy to recognize the wear of the first spot and subsequently of the other two spots, thus showing the feasibility of this technique, while a quantitative analysis of the data and a comparison with the other methods are in progress.

5. Conclusions

It has been shown that pure radioactive beams with intensities of several ppA can be produced by the offline activation technique at small accelerators and that their use can yield valuable information in both basic and applied physics. The possibility of customizing the depth profile, controlled by the incident energy, offers interesting perspectives in the determination of submicrometric wear control. The good magnetic optics properties of the beam and its intensity, in conjunction with a windowless gas target, will allow the investigation of ^{7}Be induced reactions on H and He of astrophysical interest.

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