PRODUCTION OF MINIATURIZED ^{72}Se/^{72}As POSITRON GENERATORS FOR APPLICATIONS IN MATERIALS SCIENCE

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Abstract

Positron lifetime measurements using β^+-γ-coincidence is a promising tool for non-destructive testing of materials [1]. Mobile equipment for the application of this technique requires miniaturised collimated positron sources emitting positrons of high energy, such as those emitted by ^{72}As (2.5 and 3.3 MeV) in order to probe bulk properties of the material. The present paper describes the production of miniaturised ^{72}Se/^{72}As positron generators fulfilling these requirements. ^{72}Se was produced by the reaction ^{70}Ge(α,2n)^{72}Se by irradiating a Ge disc with 38 MeV α-particles. The positron generator was then produced by a vapour deposition process. By melting the irradiated Ge disc in a closed graphite crucible with a fine nozzle in the top cover a Se vapour jet was produced that allowed the deposition of Se vapour inside a cylindrical bore of 0.6 mm diameter in a gold cylinder of 2 mm outer diameter. With preliminary process parameters ^{72}Se/^{72}As positron generators of 0.6 mm diameter and about 0.5 to 1 MBq positron activity have been produced in 1 hour of vapour deposition. About 20% of the ^{72}Se activity produced in the Ge was recovered. The process could be further optimised if required.

INTRODUCTION

Recently Holzwarth and Schaaff [1] demonstrated the utility of positron-lifetime measurements for non-destructive monitoring of fatigue-damage evolution and residual lifetime assessment of austenitic stainless steel specimens subjected to symmetric push-pull cyclic deformation. The sensitivity of positron annihilation to fatigue damage is due to the increase of vacancy-like defects in the material that act as positron trapping sites (e.g. [2]). Trapping sites exhibit a locally lower electron density. Therefore trapping reduces the annihilation probability with an electron and prolongs the lifetime of the positron. Consequently, an increasing average positron lifetime indicates an increasing defect density, i.e. accumulation of damage. The experimental method [3] requires positron sources of less than 1 mm diameter and positron energies that enable a sufficient penetration into the material in order to probe bulk properties. Positron annihilation measurements are performed by β^+-γ coincidence measurements, with the two signals created by the passage of a positron through a 1 mm plastic scintillator, as outlined in Fig. 1, and by the detection of a 511 keV annihilation photon by a γ-detector.

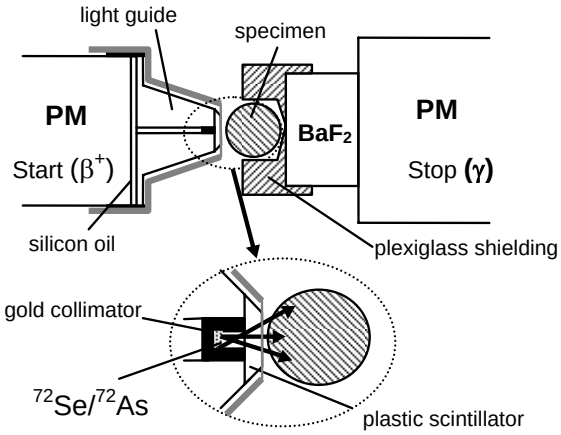

Figure 1: Experimental set-up for the positron annihilation measurements (for details see [3]). The positron generator is deposited inside a gold collimator.

For its high positron energies ^{72}As has been selected; it decays to ^{72}Ge by two specific routes, emitting positrons with energies of 2.5 or 3.3 MeV (see Fig. 2).

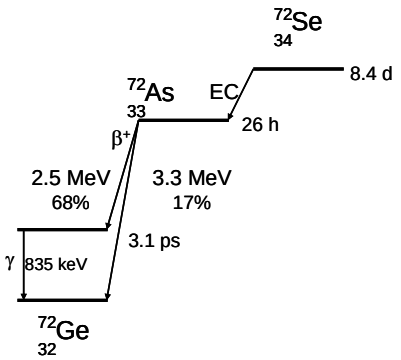

Figure 2: Decay scheme of ^{72}Se

The location of the start scintillator between specimen and source leads to a reduction of invalid annihilation events since only those positrons that are emitted in the right direction produce a start signal. The positron energy is reduced by about 150 keV for approximately perpendicular passage through the thin plastic scintillator. By discriminating electronically against the higher energy losses caused by positrons passing the plastic scintillator transversely, coincidence events associated with positrons travelling at an angle to the sample surface are reduced.

The positron generator is integrated into the assembly by removing the Plexiglas light guide from the photo multiplier tube and inserting the gold β^+-source substrate

and collimating cylinder into the central channel until it is in contact with the plastic scintillator.

RADIOISOTOPE AND SOURCE PRODUCTION

The ^{72}Se has been produced by irradiation of Ge discs of natural isotope composition of 10 mm diameter and 2 mm thickness.

The irradiations were performed using the JRC's Scanditronix MC40 cyclotron with α-particles of 38 MeV energy and a beam current of 1.5 μA on the target for 6 hours. The beam current was limited by the indirect cooling of the germanium disc.

About 92 hours after end of bombardment (EOB) the Ge disc was manually removed from the target holder and transferred to the vapour deposition device as shown in Fig. 3 and 4. The device has been designed to deposit ^{72}Se in a bore of 0.6 mm diameter in a gold cylinder of 2 mm outer diameter and 5 mm length which acts as source substrate and collimator. For the latest experiments the depth of the bore has been reduced from 1 mm to 0.3 mm in order to facilitate Se deposition on the deepest point. Collimation was then achieved by a 1 mm thick ring of gold with 2 mm outer and 0.6 mm inner diameter that has been inserted in the Plexiglas together with the positron generator. This assembly method also reduced uncollimated positrons being emitted from ^{72}Se deposited on the outside border of the bore.

The irradiated Ge disc was inserted into a graphite crucible and closed with a cover with a graphite nozzle of 0.3 mm diameter (see detail in Fig. 4). Above the crucible the gold cylinder was clamped in a copper cooling finger by use of a tantalum ring. The face of the tantalum ring towards the crucible was polished for reflecting as much IR radiation as possible in order to facilitate cooling. Tantalum was chosen because its thermal expansion is lower than that of copper and assures good clamping when heating up the assembly. For easy removal and further handling of the positron generator (gold cylinder) the copper cooling finger consisted of two conic parts, the one directly water cooled, the second one, removable, holding the gold cylinder, was blocked in his position by two pins (see Fig. 4). In order to facilitate handling and integration of the gold cylinder in the detector-source assembly a small wire was soldered on its back side.

After evacuation of the chamber to about 10^{-6} mbar the gold cylinder and the graphite nozzle were manually aligned with an x-y-z micrometer screw positioning system under optical control using two pyrometers with magnifying optics under an angle of 90 degrees. The axial alignment was better than 0.1 mm and the distance between nozzle and gold cylinder was about 0.5 mm. During heating a slight adjustment of the z-distance was required due to the thermal expansion of the graphite crucible and its supporting structure.

The crucible was electrically heated by a tantalum heating conductor coil. During generator production the crucible and the graphite nozzle were kept at a temperature of 1050°C for 1 hour. The temperature was measured with the pyrometers.

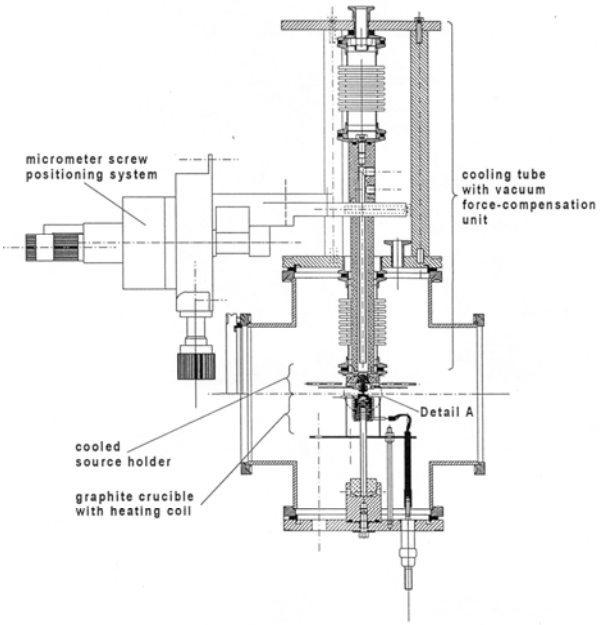

Figure 3: Vapour deposition facility for the production of ^{72}Se/^{72}As positron generators. Not shown is the vacuum part on the right side as well as the two pyrometers with magnifying optics for temperature measurement and control of the mechanical alignment.

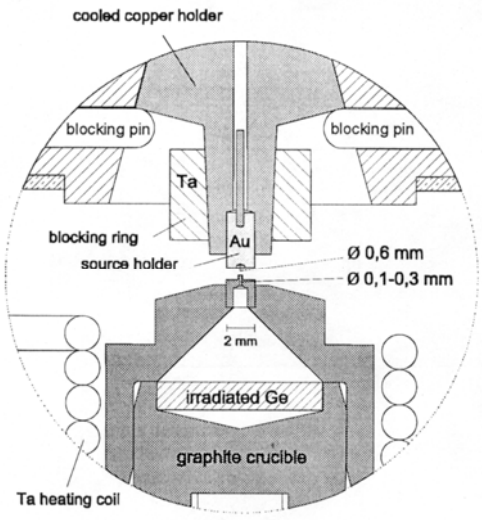

Figure 4: Detail A of Fig. 3 showing the position of the Ge disc in the crucible before melting, the nozzle on the top of the crucible cover and the fixation of the gold cylinder as substrate for the positron generator.

Upon reaching its melting point, the Ge disc transforms into a sphere due to its high surface tension. The shape of the crucible kept the sphere in a centred position. Fig. 5 shows the vapour pressure curves of Ge, Se, As and graphite as a function of temperature. At the melting point of the Ge the vapour pressure of Se is 10^{10} times higher than that of Ge. In view of the high diffusivity of Se in liquid Ge the atmosphere in the crucible is expected to consist only of Se and some As vapour. Under these circumstances the efficiency of the overall deposition process is determined by the escape probability of the atoms through the nozzle.

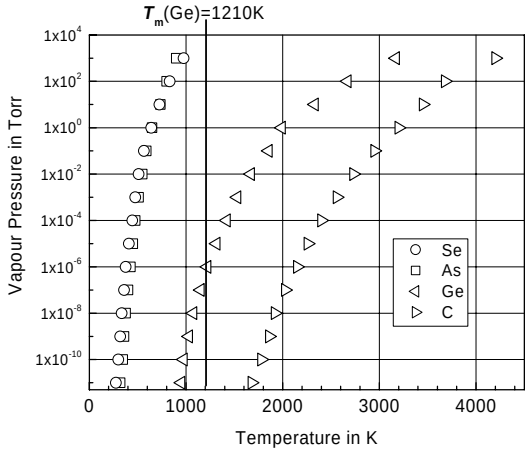

Figure 5: Vapour pressure of graphite, Ge, Se and As as function of temperature according to [4]

RESULTS

With the present irradiation parameters (E_α=38 MeV, I_α=1.5μA, t=6h) calculations indicate that about 7.5 MBq ^{72}Se should have been produced. Qualitative γ-spectroscopy performed 24 h after EOB on the Ge disc revealed the presence of the ^{72}As (from ^{72}Se), ^{73}Se, and minor quantities of ^{75}Se and ^{74}As. In order to allow the decay of short-lived undesired activation products the generator was usually produced about 92 h after EOB. 4 hours after the end of the vapour deposition process the assembled generator-detector unit typically exhibited an activity of 150 kBq of ^{72}As and about the same activity of ^{75}Se. In spite of its half-life of 17.8 d, ^{74}As could only be detected in traces. ^{73}Se had effectively decayed away completely. This indicates that Se is preferentially deposited presumably because the Se vapour consists of single atoms whereas the As tends to form As_2 and As_4 molecules [4] which reduces the probability of As to pass the nozzle. The development of the activity versus time (A_2(t)) of ^{72}As is described by Eq. 1.

$$A_2(t) = N_1(0)\lambda_1\lambda_2\left(\frac{\exp\{-\lambda_1 t\}-\exp\{-\lambda_2 t\}}{\lambda_2-\lambda_1}\right) \quad (1)$$

where N_1 denotes the number of ^{72}Se atoms, and λ_1 and λ_2 are respectively the decay constants of ^{72}Se and ^{72}As that can be calculated from the half-life data in Fig. 1.

Fig. 6 shows the evolution of the coincidence count rate during the positron lifetime measurements. This should be directly proportional to the ^{72}As activity since the solid angles for positron emission and 511 keV photon detection as well as the detection probabilities are constant. The curve can nicely be fitted by Eq. 1 and indicates no significant presence of ^{72}As at the end of vapour deposition. In order to estimate the efficiency of the process it is assumed that the 150 kBq of ^{72}As measured 4 h after deposition emerge from the decay of ^{72}Se. This means that about 20% of the ^{72}Se content of the Ge disc was recovered.

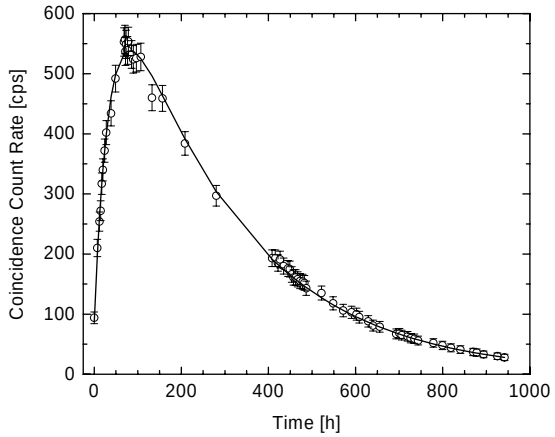

Figure 6: Development of the coincidence count rate in the positron lifetime measurement (line: fit to Eq. 1).

CONCLUSIONS

Point-like metallic ^{72}Se/^{72}As positron generators can be produced by vapour deposition from irradiated Ge discs. About 20% of the activity produced by the reaction ^{70}Ge(α,2n)^{72}Se can easily be recovered. The process is highly selective and minimizes co-deposition of As isotopes. The process parameters (temperature, duration) could be further optimized.

REFERENCES

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