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PRODUCTION OF ^{22}Na TARGETS OR POSITRON SOURCES BY MEANS OF ION IMPLANTATION

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ABSTRACT

The production and properties of high-purity ^{22}Na samples prepared by means of implantation with an electromagnetic mass separator are reported. The obtained 0.7 - 3.0 mCi radioactive ^{22}Na sources were successfully used as accelerator targets to study for the first time the resonances in the astrophysically important $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ reaction. It is shown that the method allows production of samples up to 30 mCi/cm², which due to their thickness of 10 - 50 µg/cm² are also ideally suited as positron sources.

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1. Introduction

During the last ten years there has been an increasing interest in compact isotopically pure samples of the radioactive ^{22}Na isotope, the purpose of which ranges from intense positron sources to accelerator targets for nuclear-reaction studies. Typically required quantities are 10 mCi to 1 Ci.

A number of methods have been investigated for concentrating and depositing the sodium activity. The conventional chemical deposition from aqueous solution [1,2,3], generally used for commercial sources, may achieve very high activities but has the disadvantages of relatively low specific activity due to ^{23}Na present as impurity, uncontrollable accompanying foreign matter, need for encapsulation, large positron self absorption and production of large amounts of liquid waste. As an alternative to the deposition from aqueous solution, some groups [4,5] have studied vacuum sublimation directly from an irradiated Al target. This method has the advantage of producing very little radioactive waste, gives high specific activities and, depending on the purity of the Al, yields very clean samples. A contamination of ^{23}Na cannot be avoided since it is produced more abundantly than the ^{22}Na during the activation of the Al. It is in principle a powerful method but the authors do not report on the production of any usable source or target.

Separation of radioactive nuclei from a target by means of vacuum sublimation has in recent years been extensively used in conjunction with on-line mass separators [6], and in particular the kinetics of the release of alkali metals from a number of metal targets have also been studied [7,8]. In this context, new rapid radiochemical separation techniques for most elements have been developed. These not only assure an efficient chemical isolation but also an isotopic separation and produce the product in the form of an energetic ion beam. This allows precision implantation of the high-purity radioactive nuclei into almost any substrate, which may be chosen to withstand heavy beam loads without further encapsulation. The art of target preparation by implantation with an electromagnetic isotope separator dates back several decades [9]. Its transfer to on-line mass separators [10] has already served to make radioactive targets for neutron-capture studies [11], and due to their small thickness, such targets are also useful as intense low-energy positron sources [12].

In this paper we report on the use of the ISOLDE mass separator [8] at CERN for the production of up to 3 mCi, 1.8×10^{16} atoms/cm², targets of ^{22}Na which have been successfully used to study resonances in the $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ reaction [13], of importance in the astrophysics scenario of ^{22}Na destruction in the hot NeNa chain [14]. The experimental study of this reaction requires a target of ^{22}Na and the detection of capture γ -rays in the presence of the intense radiation from the target material itself. In a previous experiment [3], the ^{22}Na was obtained commercially in the form of an aqueous $^{22}\text{NaCl}$ solution, which was dried onto a Ta backing. This target (60 μCi) could withstand only moderate beam currents, and was also found to contain significant amounts of stable contaminants such as Na, Mg, Si, Cl and heavier

elements. In spite of substantial efforts, only upper limits on the strengths of potential resonances were reported [15]. In the following we report only on the "key" to these studies, i.e. preparation of the ^{22}Na target. Other aspects of the work are described elsewhere [13].

2. Production of ^{22}Na implanted targets

An optimum ^{22}Na target will have a radioactivity of about 10 mCi ($\approx 4 \times 10^{16}$ atoms). In a target spot of 0.5 cm^2 , this amount of ^{22}Na would yield (at a proton energy of $E_p = 1 \text{ MeV}$) a target thickness of 0.6, 1.5 and 2.4 keV for a pure ^{22}Na target, a $^{22}\text{NaCl}$ and a $^{22}\text{NaTa}$ target, respectively. In order to maximize the capture γ -ray yield in the $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ studies, the ^{22}Na target stratum should be as pure as possible and the target thickness should be greater than the beam-energy spread ($\xi \leq 600 \text{ eV}$). In order to minimize the background contributions from reactions on the target substrate the ^{22}Na should be implanted into (chemically pure) Ta or other high-Z backings which are particular stable against high beam loads.

The substrate to be used for the ion-beam implanted ^{22}Na targets was investigated [16] separately by implantation of ^{23}Na ions with doses above $10^{17} \text{ atoms/cm}^2$ in Ni, Zr, Nb, Ta, and Au backings at $E_{\text{Na}} = 30$ and 50 keV . It was found that the requirements of a high ^{23}Na concentration, a homogeneous ^{23}Na distribution in the backing, and sufficient stability under high beam power were best fulfilled by Ni. The Ni layers were also found to be sufficiently free of contaminants.

The implantation of the ^{22}Na was carried out at $E_{\text{Na}} = 60 \text{ keV}$ by means of the ISOLDE-II on-line mass separator at CERN, which is specially equipped to handle large amounts of radioactivity. In the on-line mode this system gives secondary beam intensities which allow collection of mCi to Ci sources for a number of nuclei [17] with half-lives of up to one month. However, on-line production of the ^{22}Na isotope is not sufficiently high for collection of the required samples for the present purpose. Instead about 35 mCi ^{22}Na (activity in July 1989) in 5g Al host material was purchased from the Los Alamos National Laboratory. This activity was made by 800-MeV proton-induced spallation reactions in an Al disc, from which 3-10 mm chips were cut. They contained, in addition to ^{22}Na nuclei, other light nuclei (such as ^{23}Na), either produced via the spallation reactions or present originally as contaminants in the Al disc. By replacing this material in the on-line target position and heating it above the Al melting point, the ^{22}Na and possibly other elements were evaporated. The released atoms flow into a surface-ionisation ion source, which lets only alkali ions (Li^+ , Na^+ , K^+ , etc.) emerge as ions, to be accelerated. With the analyzing magnet of the separator, the ^{22}Na beam may be selected for implantation in the Ni backing.

A standard ion-source system (Fig. 1.) with a tungsten-surface ionizer of 0.5-mm outlet diameter [18] was used for the separation, with a mass resolution of $M/\Delta M = 1000$. In order to test the efficiency of the system, a dummy separation was first made with 20 nCi ^{24}Na (induced in 0.5 g Al of 99.9% purity by means of the 600-MeV proton beam from the CERN synchrocyclotron). The irradiated Al piece contained in an Al_2O_3 boat was placed in the Ta

container of the surface-ionisation unit and mounted on the ISOLDE test separator, which has the same specifications [6,8] as ISOLDE-II. By slowly raising the temperature of the container to about 750°C, the ^{24}Na was released into the ion source and accelerated. From the measurement of the collected ^{24}Na activity an efficiency of $56 \pm 5\%$ for the ^{24}Na separation was found. About 5% of the activity was left on a graphite plug inside the Ta container and more than 8% of the activity was found at the extraction electrode. Less than 5% was left in the Al and Al_2O_3 boat.

With this efficiency and the 35 mCi material available, it was planned, at the ISOLDE-II facility, to produce two ^{22}Na targets with about 10 mCi each. Preliminary experiments on blank Ni backings showed that the known (p,g) resonance yield induced on the ^{58}Ni and ^{60}Ni isotopes degrade the signal-to-background ratio in the planned experiments above $E_p = 1$ MeV. Thus, one backing was a Ni-Au-Ta sandwich, which consisted of a thin (20 mg/cm²) Ni layer (but thick enough to stop a 60-KeV Na beam) evaporated onto, a 40-mg/cm² Au layer (of proton-stopping thickness for $E_p \leq 1.5$ MeV) on a 0.2-mm-thick Ta backing. Although this sandwich target has a reduced (p,g) yield from the Ni isotopes, it was found that it could not withstand high beam loads: after a few coulomb of bombardment, the Ni layer started to blister. Consequently, such a ^{22}Na target was planned for use in the $^{22}\text{Na}(\text{p,g})^{23}\text{Mg}$ studies only at $E_p \geq 1$ MeV, with moderate beam currents. At lower energies, where the Ni background yield is negligible, a pure Ni layer of proton stopping thickness (40 mg/cm²) on a 0.2-mm-thick Ta backing should be used. Such targets were found [16] to be rather stable against high beam powers.

The first substrate to be doped with ^{22}Na was the Ni-Au-Ta sandwich. When the ^{22}Na source material was loaded into the Al_2O_3 boat, it was inspected visually for the first time. The Al pieces had a blackish surface layer suggesting significant contamination. Indeed, when the material was placed into the surface ionization unit and the oven was heated up to the standard temperature, the material outgassed strongly. As a consequence, the ionization efficiency deteriorated significantly, down to about 10%. The implantation into the Ni-Au-Ta substrate was stopped after an activity of about 3 mCi was accumulated. The second backing, the Ni-Ta target, was then implanted with the remaining material left in the ion-source system, which produced a target of about 0.7 mCi activity.

3. Properties of the ^{22}Na implanted Ni-Ta target

Although the Ni-Au-Ta target had nearly a factor of 4 higher activity (on a similar area) than the Ni-Ta target, the $^{22}\text{Na}(\text{p},\gamma)^{23}\text{Mg}$ resonance yield was actually lower than that from the Ni-Ta target, for reasons not understood. Therefore all data reported below were obtained with the latter target.

The ^{22}Na activity of the Ni-Ta target was measured with a 7.6-cm diameter x 7.6-cm NaI(Tl) crystal at a distance of 190 cm using the 1.27-MeV γ -ray: 0.69 ± 0.07 mCi [13]. This activity corresponds to a total number of ^{22}Na nuclides of $N_{\text{tot}}(^{22}\text{Na}) = (2.90 \pm 0.30) \times 10^{15}$.

This activity measurement was repeated several times after the target was bombarded with known accumulated charge. The results (Fig. 2a) show that the target was quite stable, i.e., even after an accumulated charge of 140 coulomb, the activity loss was only 35%.

The distribution of the ^{22}Na nuclides in the target was determined by scanning the γ -activity ($E_\gamma = 1.27 \text{ MeV}$) with a 2-mm-wide slit in a 10-cm-thick lead collimator in front of a Ge detector. Figs. 2b and 2c show the resulting activity distributions measured along two orthogonal directions (x,y). The dotted curves are calculations [13] assuming a homogeneous distribution of the ^{22}Na nuclides over an area of radius $R_x = 4.0 \text{ mm}$ and $R_y = 4.5 \text{ mm}$. Obviously, the assumption is fairly well fulfilled for the central region of the target, while there are additional ^{22}Na nuclides outside this region ($\approx 15\%$) due to the "tail" of the ^{22}Na ion-beam. Since the beam-defining collimator had a 4.5-mm radius and the proton beam had an effective radius of $R_p \approx 2.5 \text{ mm}$, a mean radius of $R = 4.25 \pm 0.25 \text{ mm}$ was assumed for the central target region, leading to a central target density of $N_a(^{22}\text{Na}) = (4.3 \pm 0.5) \times 10^{15} \text{ nuclides/cm}^2$. As for the target-activity measurements, the target was re-scanned after proton bombardment. The results showed that predominantly the central region of the target had deteriorated, over an area consistent with the beam size.

The effective target thickness, $\Delta_{\text{eff}} = 9 \pm 1 \text{ keV}$, observed for the $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ resonance at $E_p = 613 \text{ keV}$ (Fig. 2d), together with the number of active ^{22}Na target nuclei, $N_a(^{22}\text{Na})$ leads to the effective stopping power, $\epsilon_{\text{eff}} = \Delta_{\text{eff}}/N_a(^{22}\text{Na}) = (2.1 \pm 0.3) \times 10^{-12} \text{ eV cm}^2 \text{ atom}^{-1}$. Assuming that the number of inactive target atoms, N_i , arises predominantly from the Ni backing, the effective stopping power is given by $\epsilon_{\text{eff}} = \epsilon_{\text{Na}} + x\epsilon_{\text{Ni}}$, where x is the stoichiometric ratio, $x = N_i/N_a$. With the stopping-power tables [19] one finds $x = 130$, i.e., one ^{22}Na nuclide per 130 Ni nuclides in the ^{22}Na target region (Table 1). The measurement of the yield curve at the 613-keV resonance was repeated after several irradiations of the target to monitor its stability. The results confirmed the loss of ^{22}Na activity (Fig. 2a) and showed also that the effective target thickness Δ_{eff} stayed nearly constant (within 10%).

From the measurement of excitation functions [13], the target contaminants ^{12}C , ^{13}C , ^{19}F , ^{23}Na and ^{27}Al were identified and found to be located mainly on the target surface. Their number densities, estimated from known resonance strengths, are summarized in Table 1. The largest contamination is carbon, arising probably from carbon deposition during the ^{22}Na implantation, when the vacuum near the target was only about $5 \times 10^{-6} \text{ Torr}$.

4. Summary and discussion

We have produced 0.7 - 3 mCi high-purity ^{22}Na targets by means of an electromagnetic mass separator. This implantation technique, in conjunction with novel and standard γ -ray detectors led to the first successful study of the resonances in the $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ reaction. This experiment involves the shortest-lived target nucleus used to date to actually observe capture γ -rays in (p, γ) reactions. Limited by sputtering, the method as described allows production of ^{22}Na targets or positron sources with $N_a(^{22}\text{Na}) \leq 10^{17} \text{ atoms/cm}^2$ i.e. 30 mCi/cm². Higher

densities may be obtained by lowering the implantation energy. In order to assure the maximum ion-source efficiency, high-purity Al should be freed from ^{23}Na and dissolved gases by prolonged vacuum melting before the activation.

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Table 1
Composition of the ^{22}Na -target implanted in Ni-Ta.

Element	Reaction	E_p (keV)	ω_γ (eV)	N^a (10^{15} atoms/cm 2)
^{22}Na	activity			4.3 ± 0.5
Ni	backing			560 ± 60
^{12}C	$^{12}\text{C}(p,\gamma)^{13}\text{N}$	457	0.64	≈ 70
^{13}C	$^{13}\text{C}(p,\gamma)^{14}\text{N}$	551	9.2	≈ 0.26
^{19}F	$^{19}\text{F}(p,\gamma)^{16}\text{O}$	341	22	≈ 0.02
^{23}Na	$^{23}\text{Na}(p,\gamma)^{24}\text{Mg}$	1023	14	≈ 0.16
^{27}Al	$^{27}\text{Al}(p,\gamma)^{28}\text{Si}$	992	2.6	≈ 0.10

a Errors of up to a factor of 2 are possible for the elements C, F, Na and Al.

Figure captions

Fig. 1: ISOLDE ion-source unit.

Fig. 2: (a) Loss of ^{22}Na activity of the implanted Ni-Ta target as function of accumulated charge on the target. (b) and (c) Two directional scans (x,y) of the ^{22}Na γ -activity distribution. The dotted curves represent calculations for a homogeneous ^{22}Na distribution with radii $R_x = 4.0$ mm and $R_y = 4.5$ mm. Also indicated are the centroids of the solid curves. (d) Excitation function near the $E_p = 613$ keV resonance of the $^{22}\text{Na}(p,\gamma)^{23}\text{Mg}$ reaction which leads to an effective target thickness of $\Delta_{\text{eff}} = 9 \pm 1$ keV.

Figure 1

Figure 2