PROPOSED NEW REACTOR-ACTIVATED POSITRON SOURCE FOR INTENSE SLOW e+ BEAM PRODUCTION

M. SKALSEY and J. VAN HOUSE

Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan 48109, USA

A novel method is suggested for producing a new positron (e+) emitting isotope in a nuclear reactor with application to slow e+ beams. The initial radiated sample is ^{124}Xe which is transformed to ^{126}I by two neutron absorptions and an intermediate decay. Over 25 Ci of positrons with a specific activity of 25 Ci/gm can be produced by this technique, allowing the generation of a slow e+ beam of over 4×10^7 e+/cm^2–s. As discussed in the conclusion, specific activities approaching 200 Ci/gm should be possible. Two designs for activation cells are presented, one with Xe in the gas phase, the other with solid Xe. Both designs allow the easy separation of the ^{126}I from other contaminants, permitting the production of a pure, high specific activity source.

1. Introduction

The process of moderating fast e+ (≤ 1 MeV) to low energy (~ 1 eV) and forming beams of "slow" e+ has been under investigation for almost two decades [1]. The technique rests on the fact that when fast e+ slow down in a material, thermal diffusion of the e+ takes place. If the e+ thermalize within a diffusion length of the surface of the material, then some of the e+ will diffuse to the surface and be ejected if the work function (typically 1–2 eV) is negative, opposite to the positive work function for electrons. The fast to slow conversion efficiency is typically of order 10^{-3} for the best moderators. These ejected e+ can be accelerated and focussed, forming a slow e+ beam.

The uses of slow e+ beams have been many and varied, examples include solid state [2,3] and surface physics [4,5], fundamental studies of positronium atoms [6–8], astrophysics [9], studies of the origins of bio-chirality [10] etc. Two noteworthy extensions of the basic moderation process used to produce slow e+ beams are: i) brightness-enhancement [11] of the beams through a remoderation process and ii) production of spin polarized slow e+ beams [12,13]. Some interesting future uses of slow e+ beams are going to require higher intensity, brighter beams than are currently available. This paper will address both issues of intensity and brightness. Some of the future uses for new beams could include:

1) angular correlation and doppler studies of bulk material and surfaces [14],
2) formation of e+ e− plasmas [15],
3) Bose condensation of positronium atoms [15],
4) e+ microscopy of surfaces, defects, cracks, and solid state devices [11,16],

5) laser spectroscopy of positronium states [7],
6) formation of anti-hydrogen [17] atoms ($\overline{H}_0$), from stored anti-protons, and
7) production of polarized anti-protons [18] from polarized $\overline{H}_0$ which were formed from polarized e+.

(This list is not exhaustive but it is intended to show the broad scope of applications for slow e+ beams. All of the projects will benefit from increased rates and brightness in slow e+ beams.)

A typical design for a slow e+ beam is shown in fig. 1. of ref. [13]. The fast e+ are provided by decays from a radioactive source. Most slow e+ beams use sources, a noteable exception being fast e+ from pair production of bremsstrahlung from electron linacs [19]. Radioactive sources are certainly more convenient to use in most situations compared to linacs. Also, we feel that the expansion potential for sources, yielding higher intensity beams, is better than for linacs which currently give pulsed beams of up to 10^8 e+/s, compared to 10^8 e+/s for sources.

The accelerating and focussing lens elements of most slow e+ beams are mainly electrostatic, although the use of predominantly magnetic guiding fields instead of electrostatic lens is also common [20]. Frequently, the slow e+ beams are deflected to eliminate the flux of fast e+ and reduce the source-related gamma-ray noise in experiments.

The radioactive source is critical to slow e+ production. Typical e+ energies from these sources range from several hundred keV to several MeV. The lower energy e+ are more efficient in the simple moderation process. For polarized beams, however, an energy of 500 keV −1 MeV is optimal. There are many requirements placed on the physical nature of these sources. The sources must be sealed in the sense that the activity

Table 1
Isotopes for slow e^+ beams. The most commonly used e^+-emitting isotopes for slow e^+ beams are listed, along with the proposed ^{126}I isotope. The fifth column, S_{max}, is the maximum specific activity of positrons per gram for isotopically pure material, producing such a sample would require mass separation techniques. The final column is the brightness-optimized deposition density obtained from S_{max} (see appendix).

Isotope	$T_{1/2}$	$\%\beta^+$	$e_{\beta^+}^{max}$ (keV)	S_{max} (Ci of e^+/g)	ρ_d^{max} (Ci/cm^2)
^{22}Na	2.60 yr	90	545	5.62×10^3	135
^{58}Co	70.8 d	15	474	4.76×10^3	98
^{64}Cu	12.7 h	19	656	7.30×10^5	21 700
^{68}Ge	288 d	89	1900	6.02×10^3	60
		1.3	800		
^{126}I	13.0 d	0.8	1130	7.94×10^2	36
		0.2	460		

must be stable under vacuum and bake-out. Small diameters of deposit are typically used ($\sim$ few mm) in order to obtain brighter beams. Thin windows ($\sim$ few mg/cm^2) of Ti or Ni often cover and encapsulate the activity. The isotopes that are frequently used for slow e^+ beams are listed in table 1, along with ^{126}I discussed below.

Most e^+-emitting isotopes must be produced using accelerators, rendering production of these sources relatively expensive. Two important exceptions are the reactor-produced isotopes ^{58}Co and ^{64}Cu. The latter is produced by thermal neutron absorption on stable ^{63}Cu, the former by the fast neutron reaction (n, p) on stable ^{58}Ni. Chemical separation of ^{58}Co and ^{58}Ni is used to increase the purity of the ^{58}Co sources. Of course, this is impossible for the ^{64}Cu production scheme, but isotopic enrichment has been suggested to increase the ^{64}Cu specific activity [21]. A slow e^+ beam facility based on ^{64}Cu has been located at the Brookhaven reactor [14] with source activities in the range of 1–10 Ci of e^+ having been achieved. The production of very intense ^{58}Co sources is being investigated at Chalk River National Laboratory and Idaho National Engineering Laboratory [22].

The purpose of this paper is to suggest a new reactor-produced isotope, ^{126}I, for use with slow e^+ beams. This isotope is produced by a circuitous route, involving two neutron activations and an intermediate decay, starting with ^{124}Xe. There exists in nature only one other analogous cycle, starting with ^{130}Ba, which appears less-promising for e^+ production. Two specific designs for xenon–iodine activation cells, presented below, have been tailored to the 2 MW Ford research reactor here on the University of Michigan campus.

2. The xenon–iodine activation cycle

Most simply stated, the reactor neutrons activate a sample, the product of the activation is also exposed to

Table 2
Cross sections for (n, γ) reactions. The neutron activation cross sections for several relevant isotopes are presented [24]. Cross sections are for thermal neutron absorption (σ_{ther}) and resonance integrals (σ_{fast}).

Isotope	σ_{ther} (b)	σ_{fast} (b)
^{124}Xe	122	3.0×10^3
^{125}I	900	14×10^3
^{126}I	9×10^3	–
^{127}I	6.2	150
^{126}Te	1.0	10
^{125}Te	1.6	20
^{126}Xe	2.3	34
^{128}Xe	4.4	330
^{129}Xe	2.0	250

the neutron flux, causing another activation. The final, second-order product is the desired e^+-emitting isotope. The initial sample is stable ^{124}Xe, whose natural abundance is 0.096% of normal Xe. Samples of xenon are available commercially [23] which are enriched in ^{124}Xe to as high as 50%. However, for our purposes, the more financially attractive 20% enrichment is assumed.

Reactor neutrons incident on ^{124}Xe produce ^{125}Xe

Table 3
Decay data for xenon–iodine cycle isotopes

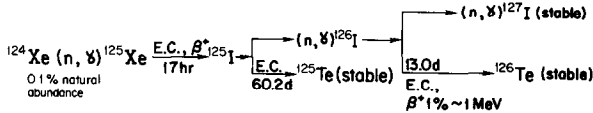| Isotope	$T_{1/2}$	Decay	Frac. (%)	E_{β}^{max} (keV)	γ-rays
^{125}Xe	17.3 h	E.C.	99.7		3% 1 MeV
		β^+	0.3	470	
^{125}I	60.2 d	E.C.	100		35 keV
^{126}I	13.0 d	E.C.	53		4.6% 1.4 MeV
		β^-	46	~ 900	
		β^+	0.8	1130	
		β^+	0.2	460	

Fig. 1. The xenon–iodine activation cycle. Seven isotopes and six processes, decays and neutron absorptions, comprise the xenon–iodine activation cycle. The initial sample material is ^{124}Xe, isotopically enriched to 20%. The desired final e^+-emitting product is ^{126}I.

Table 4
The physical properties of the elements important in the xenon–iodine activation cycle

	ρ-density	Melting point (°C)	Boiling point (°C)
Iodine I$_2$	4.93 g/cm^3	114	184
Xenon Xe	5.9 g/l (gas) 2.7 g/cm^3 (solid)	−112	−108
Tellurium Te	6.1 g/cm^3	452	1087

through the (n, γ) reaction. The cross sections for this and the other processes are shown in table 2. The decay of ^{125}Xe has a half-life of 17.3 h and produces ^{125}I, which has a much longer half-life (60.2 d). After a few days of activation, most ($> 90\%$) of the $A = 125$ radioactive atoms are ^{125}I. Therefore, ^{125}I can be used as a second sample for neutron activation, the (n, γ) reaction yielding ^{126}I which is the e^+-emitter of interest. This entire sequence of isotopes, along with deleterious side chains, is depicted in fig. 1. Pertinent nuclear data on the decay of these isotopes is presented in table 3.

The final neutron activation shown in fig. 1, ^{126}I(n, γ)^{127}I, is of particular importance. This step depletes the ^{126}I isotope, which is the desired product. The known cross sections for this reaction (table 2) indicate the major sensitivity is to thermal neutrons. It is possible to use a Cd shield to significantly reduce the thermal neutron flux but not the fast neutron flux. The previous neutron activation shown in fig. 1, ^{125}I(n, γ)^{126}I, would not be overwhelmingly reduced in the presence of such a shield (table 2). The same is true of the first neutron activation, ^{124}Xe(n, γ)^{125}Xe, shown in fig. 1. This shielding technique will be employed in the two designs presented in the next section.

3. Two cell designs for the Ford nuclear reactor

The 2 MW Ford Nuclear Reactor, commissioned on the University of Michigan campus in 1957, is capable of irradiating small samples in the core with a maximum thermal neutron flux of 3×10^{13} n/cm^2 s. The proposed location for activation has a thermal neutron flux of 2×10^{13} n/cm^2 s and a 5 cm diameter cell can be accommodated. The fast neutron flux at this position is one-third the thermal neutron flux [25], that is, the total activation rate, dN/dt, using the cross sections σ_t and σ_f in table 2, is given by

$$dN/dt = \Phi_t \left(\sigma_t + 1/3\sigma_f \right) M/A,$$

where Φ_t is the thermal neutron flux, M is the mass of the sample being activated, and A is the atomic weight.

Two different designs for activation cells are proposed. One is based on the gas phase for the Xe target material, the other is based on the solid phase. The bulk

physical properties of the Xe target material, of the I$_2$ product material, and of the Te contaminant, are shown in table 4. Two important points can be discerned from table 4. First, Xe can be solidified at liquid nitrogen (LN$_2$) temperature. Second, it is possible to separate I$_2$ from Xe in the gas phase simply by condensing the I$_2$. In the gas phase cell design, presented next, this separation is performed while the activation is occuring.

Fig. 2 depicts an activation cell designed to be lowered into the core of the Ford Reactor. Approximately 7.5 g of Xe gas, enriched to 20% of ^{124}Xe, is in the cell at a pressure of 2.5 atm. The gas is allowed to flow freely between the Xe gas chamber and the I$_2$ condensation chamber. The walls of the Xe chamber are maintained at 200–300°C to prevent I$_2$ from condensing. A 3.8 cm diameter tungsten (W) disk in the I$_2$ chamber is water-cooled through the control cable, which also carries electrical power for heaters and thermocouple and pressure readouts. The ^{125}I produced
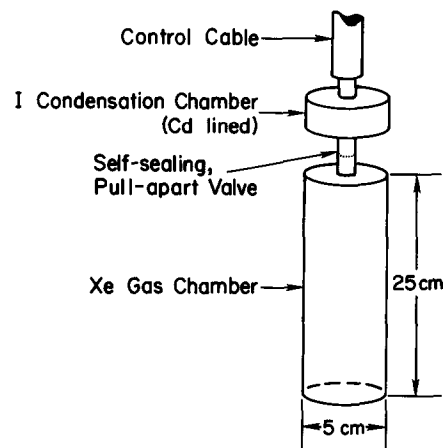

Fig. 2. Gas phase activation cell. Thermal and fast neutrons activate Xe gas, enriched in ^{124}Xe, which circulates throughout the cell. ^{125}I collects in the upper chamber and is activated by fast neutrons. To retrieve the e^+ source, the upper I$_2$ chamber is removed from the reactor leaving the Xe chamber in the core.

from the Xe activation condenses on one surface of the W disk. Since the I_2 chamber is lined with a Cd shield to absorb the thermal neutrons, the activation of ^{126}I will occur primarily with fast neutrons. As mentioned earlier, the purpose of the Cd shielding is to prevent the depletion of the ^{126}I product by thermal neutrons.

A sample schedule of operation might be: 1) Irradiation of the cell for two weeks, at which time the I_2 chamber is pulled out of the reactor, leaving the Xe chamber in the core. 2) The W disk with the ^{126}I is placed in the slow e^+ beam and two weeks of counting can commence. (Note the e^+ source intensity will drop by a factor of two during this period.) 3) Another W disk is put in the I_2 chamber which is then re-attached to the Xe chamber in the reactor core and activation begins again. 4) After two weeks, the two W disks are interchanged and a repeating cycle ensues. It is not clear how many times the W disks can be recycled and still maintain a high specific activity deposit. This will depend on the amount of Te contamination and on the mobility of the I_2 on the disks. If five recyclings are possible before new W disks are needed, an average of 2.5 Ci of e^+ source activity (250 Ci of ^{126}I) will be obtained over the life of the disks.

Another possible activation cell design could be based on solid Xe at LN_2 temperature surrounded by a Cd thermal neutron shield. In this case, it would not be possible to separately shield the I_2 and not shield the Xe from thermal neutrons, since the I_2 cannot be separated from the Xe while the activations are occurring. But this is unimportant, since no significant depletion of ^{126}I occurs from fast neutrons and the production rate is hardly affected. However, a much more massive Xe sample, resulting in a higher final ^{126}I activity, can be placed in the available space in the reactor core. For example, a 75 g solid Xe sample is 27.8 cm³ (a 3.8 cm diameter ×2.5 cm cylinder). If the enrichment of the ^{124}Xe is 20% then the expected average activity is 25 Ci of e^+ (2500 Ci ^{126}I). Separation of the I_2 from the Xe would occur after the sample had been removed from the reactor. Techniques similar to those used in the gas phase activation cell would be used to permit a separation after the Xe sample had slowly warmed and evaporated.

4. Discussion and conclusions

In either cell design, the specific activity of e^+ of the produced source would be in the range (22–26) Ci/gm. As discussed in the appendix, the maximum deposition density, ρ_d, of an ^{126}I source at this specific activity would be $\rho_d = 1.1$ Ci/cm². This is ~ 25% of the 4.4 Ci/cm² deposition density of the ^{64}Cu source located at the Brookhaven reactor. If the ^{126}I source is used in a planar source geometry with a solid neon moderator

[26] with a 7×10^{-3} conversion efficiency, a slow e^+ beam of 3×10^8 e^+/cm² s could be produced at the Ford reactor with a ^{124}Xe sample enriched to 20%.

Substantial increases in specific activity can be obtained at other reactors because the activities scale as the square of the fast neutron flux ratio, due to the fact that two neutron absorptions are required. Thus, a fast neutron flux of $\Phi_f = 7 \times 10^{13}$ n/cm² s would yield specific activities of 200 Ci/gm for a 20% enriched ^{124}Xe sample, resulting in a deposition density of 9.3 Ci/cm², roughly twice the value of the ^{64}Cu source. Use of a solid neon moderator [26] on this last source would give, for a planar source geometry, a slow e^+ beam current density of 2.5×10^9 e^+/cm² s. The relation using the fast flux ratio squared assumes that the shielding of the samples from the thermal neutron flux remains effective. A thermal neutron flux of about 10^{14} n/cm² s will deplete the ^{126}I as fast as its decay and would result in significant losses in activity.

A number of advantages of the ^{126}I source exist:

1) High specific activities of the ^{126}I source can be achieved, resulting in beams of up to 2.5×10^9 slow e^+/cm² s.

2) Its long (13 day) half life would be more convenient for certain types of experiments.

3) The costs of maintaining a continuous supply of ^{126}I are modest, due to relatively simple and infrequent handling procedures for producing the sources [28] and a low (4%/month) depletion of the enriched ^{124}Xe sample.

4) Its ease of separation and installation into a beam, and its ease of deposition onto unusual geometries are additional advantages.

5) High specific activities (25 Ci/gm) can be obtained at the type of small research reactor available at many locations around the world, allowing the production of a number of intense slow e^+ beams.

Preliminary experiments are being planned to test the ideas put forward in this paper.

Appendix: Source thickness considerations

Here we consider the effective source strength per unit area, ρ_d, of a source which is deposited in a thick layer, including the effects of source self absorption. This quantity, ρ_d, is important since the brightness [11] of the resultant slow e^+ beam will scale with ρ_d.

The intensity of fast e^+ which reach the surface of a source of thickness l emitted from a layer of thickness dx located a distance x from the surface of the source can be expressed as

$$dI = \rho A S \, e^{-\rho x/\mu_0} \, dx, \qquad (A.1)$$

where ρ is the mass density of the material, A is the area of the source, S is the specific activity, and μ_0 is

the mass attenuation coefficient [29] of the deposited isotope. In this expression, corrections due to the $1/r^2$ geometric attenuation are neglected; i.e., it is assumed that the source diameter is much greater than the thickness l. In addition, the fact that up to 25% of the high energy e^+ can annihilate in flight into one or two γ-rays in thick, high atomic number, absorbers [30] is ignored.

With the definition $\rho_d \equiv I/A$, and using $\mu = \rho l = I_0/SA$, where I_0 is the total source strength in Ci, integration of eq. (A.1) over the thickness yields the deposition density as a function of μ_0, μ, and S:

$$\rho_d = \mu_0 S(1 - e^{-\mu/\mu_0}). \quad (A.2)$$

The maximum value of ρ_d occurs as $\mu \to \infty$, where $\rho_d^{max} = \mu_0 S$. Using the semiempirical expression [29]

$$\mu_0 = 0.048 \left(E_{\beta^+}^{max} \right)^{1.14}, \quad (A.3)$$

where $E_{\beta^+}^{max}$ is the isotope endpoint energy in keV, and μ_0, which is in mg/cm^2, is adjusted from ref. [29] to account for the 20% greater range [31] of e^+ over e^-, gives the values of ρ_d^{max} shown in table 1 for several common slow e^+ emitting isotopes, and for the ^{126}I source discussed in this paper. In practice, deposition densities approaching ρ_d^{max} can be achieved for $\mu \simeq (2-3)\mu_0$.

We parenthetically note that small laboratory beams are typically based on commercial ^{22}Na where $S = 500$ mCi/mg for a deposit consisting of ^{22}NaF. These sources are usually deposited in thin layers to maximize intensity. One example [13] involved deposition of 50 mCi ^{22}Na onto a 3 mm diameter spot, resulting in $\rho_d = 0.7$ Ci/cm^2. If, however, these sources could be reliably deposited without loss of specific activity in a thickness of ~ 2 μ_0, a value $\rho_d \approx 14$ Ci/cm^2 would result. Use of such a thick source would increase the brightness of laboratory based slow e^+ beams by a factor of 20 or more, without the technological complexity of brightness enhancement by remoderation [11].

Note added in proof: After this paper was accepted, we were informed by Dr. Ben Brown of Harvard University that the resonance integral for ^{126}I is $\sigma_{fast} = 5 \times 10^3$ b. This is in disagreement with table 2. Because of this correction, the use of a thick Cd shield, as proposed in this paper, is no longer as attractive. Yield and

specific activity estimates should be reduced by about 10% for the Ford Reactor activations. Reductions in the quantities presented for other, larger reactors will be as large as factors of 2–3.

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[28] Design of the handling system must take into account radiation and shielding considerations. Assume we have a 50 Ci e^+ source (5000 Ci of ^{126}I) that was just removed from the reactor. The hottest isotope is ^{125}Xe($T_{1/2} = 17$ h) which will remain in the core if using the gas phase cell or which will be removed if using the solid phase. The γ-rays from this decay will give about 200 Ci ^{60}Co equivalent. The next hottest isotope is ^{126}I($T_{1/2} = 13$ d), the desired e^+-emitter, which is definitely removed from the reactor no matter which cell is used. The γ-rays from this decay would give about 150 Ci ^{60}Co equivalent. Clearly, substantial shielding and remote handling, equivalent to that already employed at the Brookhaven ^{64}Cu source, would be required to use these sources. Another topic of concern is the release of radioactive I_2 into the environment, due to the strong interaction of I_2 with living systems. Present regulations permit maximum releases of insoluble ^{125}I and ^{126}I in air of 6×10^{-9} and 1×10^{-8} μCi/ml respectively. Cell design and handling procedures will be critically important in insuring minimal radiation releases.

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