DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

CONF-830770--2

THE GENERATION OF MONOENERGETIC POSITRONS

CONF-830770--2

DE83 015535

L. D. Hulet, Jr., J. M. Dale, P. D. Miller, Jr.,
C. D. Moak,^a S. Pendyala,^b W. Triftshäuser,^c R. H.
Howell,^d R. A. Alvarez^d

^aOak Ridge National Laboratory, Oak Ridge, Tennessee,
USA

^bState University of New York, College at Fredonia,
Fredonia, New York, USA

^cHochschule der Bundeswehr, Munich, FRG

^dLawrence Livermore National Laboratory, Livermore,
California, USA

ABSTRACT

Many experiments have been performed in the generation and application of monoenergetic positron beams using annealed tungsten moderators and fast sources of ^{58}Co , ^{22}Na , ^{11}C , and LINAC bremsstrahlung. This paper will compare the degrees of success from our various approaches. Moderators made from both single crystal and polycrystal tungsten have been tried. Efforts to grow thin films of tungsten to be used as transmission moderators and brightness enhancement devices are in progress.

INTRODUCTION

In 1974 Pendyala¹ reported spectra for positrons emitted from several different transition metals, among these were results for tungsten. In 1980 Dale, Hulet, and Pendyala² showed that tungsten could be made into a highly efficient positron moderator if it is annealed in vacuum at temperatures greater than 1500°C. Annealing accomplishes two purposes, the cleaning of the surface by the volatilization of oxides, and the reduction of defects that decrease the escape probability of the thermalized positrons. Tungsten surfaces prepared by annealing are very stable in air at room temperature, the oxide coating that forms is usually less than five monolayers. Thus, the tungsten

MASTER

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a non-exclusive, royalty-free license in and to any copyright covering this article.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

moderator is very 'practical'; in situ preparation, although probably desirable, is not necessary; the moderator can be alternately exposed to air and replaced under vacuum without serious reduction in efficiency. The annealed tungsten moderator has been adopted by a large number of other workers doing experiments with monoenergetic positrons. For example, it was used in the first low energy positron diffraction experiments, by Rosenberg, Weiss, and Canter³ Zitzewitz and co-workers⁴ have used it to obtain high yields of polarized positrons.

Since the initial discovery of the effectiveness of annealed tungsten as a positron moderator, the authors have made many attempts to improve its efficiency. Single crystal, polycrystal, and fine particle tungsten, annealed under varying conditions, have been prepared and mounted in several different orientations with respect to the fast source. We have also experimented with several different fast positron sources, and have made attempts to study the effects of altering the spectrum of the fast positron source with absorbing films of different materials. With the exception of the LINAC work, these results will be presented and discussed below. LINAC work will be presented by Howell and Alvarez and their co-workers in another paper.

RESULTS AND DISCUSSION

Annealing Procedure

Annealing of tungsten should be done at temperatures greater than 1500°C, above the melting point of tungsten oxide. This effects volatilization of the oxide and cleaning of gross contaminants from the surface. For polycrystalline material appreciable grain growth also occurs, which serves to remove defects that trap thermalized positrons and therefore increases the distance over which they can diffuse and escape from the inside of the moderator. The authors have initially reported,² that for polycrystalline material, best yields of moderated positrons were obtained when annealing was done at 2200°C. Since that time we have annealed at temperatures as high as 2600°C; yields were slightly better, but only by factors of 50% or less. Vacuum conditions for the annealing are not critical. Initial studies by the authors were done using vacuum pressures as high as 10^{-4} torr. Good vacuum conditions and high annealing temperatures appear to decrease the amount of energy loss in the moderated positron peak. The experience of others suggests that moderated positron yields are better if the tungsten is prepared in situ under high vacuum conditions. Mills⁵ reports that yields can be as high as 3×10^{-3} for single tungsten crystal moderators prepared in situ. On the other hand, Zitzewitz and co-workers⁴ report yields as high as 1×10^{-3} for polycrystal tungsten moderators that have

been exposed to air for short periods of time. The best yields obtained by the authors were about 5×10^{-4} . All of the authors' work has been done under conditions in which the moderator was exposed to air between the stages of preparation and use.

Moderator Orientation and Form

Mills⁶ has explained in detail why the backscatter arrangement is preferable for the orientation of the moderator. His positron guns are arranged such that the moderator surface is normal to the optic axis along which the slow positrons are extracted. Their sources, which are ⁵⁸Co mounted on small needles, are placed directly in front of the moderator surface. This allows the exploitation of the special emission characteristics of moderated positrons: the negative work function of the moderator causes the positrons to be ejected in a direction almost normal to the moderator surface, therefore the positrons are projected parallel to the optic axis of the gun, allowing them to be accelerated and condensed into a small spot size. Whenever thick moderators are used, and the size and shape of the fast positron source permits, the backscatter configuration is obviously the preferred arrangement. For ²²Na sources, however, such as that available to the authors, the bulkiness of the capsule makes this arrangement difficult. We were forced to perform most of our experiments with the moderator surfaces oriented parallel to the optical axis of the slow positron gun. For polycrystal moderators the vane configuration was used in most experiments. That is, the polycrystal material was cut into strips, 1-3 mm in width, and mounted as a "venetian blind" arrangement in front of the fast source. Spacings between the vanes were usually about equal to their widths. Since the moderated positrons are expected to be emitted mostly normal to the vane surfaces, they must be turned 90° in direction in order to extract them from the gun. This is obviously an undesirable situation, leading to poor brightness of the gun, but it was necessary because of the shape and size of the fast positron source.

Single crystal surfaces of tungsten, with (110) orientations have been prepared and arranged in the form of a rectangular cavity, 6 mm square, 14 mm in length and annealed at 2600°C. The ²²Na source was placed at one end of the cavity and the moderated positrons were extracted from the other end by an electrostatic field. Experimental arrangements have been shown diagrammatically in a previous report.⁷ Slow positron yields were about the same as for the polycrystalline vane moderators.

Mills⁶ has suggested that fine particle moderators, having larger surface areas, might yield larger quantities of slow

positrons. Two devices of this type, consisting of fine particles deposited on the inside of cavity configurations, have been studied. One was prepared by hydrogen reduction of tungsten hexafluoride, particle sizes ranged from 1/2-50 micrometers. Another was prepared by hydrogen reduction of tungsten oxide smoke, the average size of its particles was about two micrometers. Neither of the two fine particle moderators showed an appreciably greater yield than the polycrystal vane and single crystal cavity configurations. The reason for this is not clear. It is true that the particle sizes were much greater than the thermalized positron diffusion lengths, but one would expect the increased moderator area to have been beneficial.

The transmission moderator would appear to be the ideal configuration. It should have two major advantages; a maximum solid angle of 2π steradians for intercepting the fast positrons from the source can be obtained; secondly, as the slow positrons emerge from the front side of the moderator they should be projected parallel to the optical axis of the gun, providing maximum brightness.

Experiments with three thicknesses of tungsten transmission moderators have been performed: 25 μm , 12 μm , 2 μm . The 2 μm moderator was prepared by repeatedly anodizing and stripping the 12 μm material. All three films were annealed at about 1800°C. In all three cases yields were rather poor, the best being about 20% of that for the vane and cavity configurations. Our interpretation of these results is that the films were too thick to serve as effective moderators, and that they utilized positrons having energies corresponding to relatively small segments of the fast positron spectrum of the source. Those fast positrons that are the most effectively moderated probably have the lower energies. The thick transmission moderators that were used stopped the lower energy positrons before they reached the front surfaces, and they came from a more narrow range of the fast spectrum. We have attempted to measure the lower energy range of the fast positron spectrum from a ^{58}Co source. The upper limit of our spectrometer (electrostatic) was 2 keV. No counts above background could be detected. Apparently the nature of the surface of the fast source prevents positrons of energy lower than 2 keV from escaping.

It is still our belief that the transmission moderator is the better configuration, however. Pendyala⁸ has calculated that if the transmission moderator is the same order of thickness as the diffusion length of the thermalized positrons the yield should be equivalent to that of a back reflection configuration. We have therefore begun efforts to grow tungsten films with thicknesses of the order of the diffusion lengths of the thermalized positrons,

50-200 nm. In our initial approaches we are using the chemical vapor deposition technique. We are exposing various substrates to tungsten hexacarbonyl, which decomposes and deposits metallic tungsten at temperatures above 300°C. Tough, ductile films have been obtained for substrates such as tungsten oxide, mica and MgO. We have been unable to form coherent films on NaCl. We have not yet determined whether the coherent films are epitaxial. We have been only partially successful in floating the deposited films free of their substrates; we have yet to test whether or not they can be annealed into free standing moderators.

Experiments with ^{11}C Fast Sources

Stein and Kauppila⁹ have reported the generation of slow positrons by the bombardment of ^{11}B with 4.75 MeV protons from a dynamitron generator. The ^{11}C isotope, which is a fast positron emitter, is formed by the following reaction.

The ^{11}B target becomes self-moderating, presumably because of the build-up of carbon during proton bombardment. Miller and co-workers¹⁰ have studied this reaction for proton energies as high as 10 MeV, using the ORNL tandem van de Graaf. Because of the greater penetrating power of the higher energy particles, the production of ^{11}C was more effective; that is, the target was activated both at the high energies and also at the lower energies of the beam as it became degraded in the penetration of the target. Experiments were started using the ORNL accelerator to bombard boron carbide targets fabricated from ^{11}B and ^{13}C , both of which form positron emitting isotopes when activated with protons. Fifty millicurie quantities of activity were produced, which corresponds to the activity of the ^{22}Na source used in other experiments. An annealed tungsten moderator was used. Yields of slow positrons were very poor, more than a factor of 10 smaller than those obtained with the ^{22}Na source. Several experiments were done to check for problems such as moderator contamination; this does not appear to be the cause of the trouble. We suspect that the poor yields might be due to self absorption of fast positrons by the target. The high energy of the bombarding protons cause them to penetrate to large depths of the target before they undergo activation events. The authors have shown¹¹ that material placed between the fast positron source and the moderator causes the moderated positron to be significantly decreased.

One of the purposes of this conference is to discuss schemes for devising high intensity sources of monoenergetic positrons to

be deployed as user facilities. The ^{11}C source would seem to be attractive and practical; we would like to apprise our colleagues of the trouble we have had with this approach and advise that techniques must be devised to overcome the fast positron self absorption problems.

SUMMARY

Over the past four years our group has performed many experiments in the generation and spectrometry of slow positrons. Many appealing ideas have arisen at workshops such as this, and we have investigated several of them. We have experienced failures as well as successes. The purpose of this paper is to apprise our colleagues of both.

The annealed tungsten moderator has proven to be very useful. A large number of other workers have made use of it. It is easy to prepare and is rugged in use, being tolerant of poor vacuum conditions, and even operable in experiments with gasses.

The development of a fine particle moderator, with a greater surface area to allow larger yields, has not yet been accomplished. Whether or not this idea is sound is not clear.

The transmission moderator, if it is to work, will probably require the fabrication of thin films, less than 200 nm in thickness. Chemical vapor deposition shows promise of being a method by which this can be accomplished.

The ^{11}C fast positron source, produced by proton bombardment, has some difficulties that must be overcome before it can be used for high intensity monoenergetic positron facilities.

ACKNOWLEDGMENT

Research sponsored by the Office of Energy Research, U. S. Department of Energy, under Contract W-7405-eng-26 with the Union Carbide Corporation.

REFERENCES

1. S. Pendyala, D. Bartell, F. E. Girouard, J. Wm. McGowan, Phys. Rev. Lett. 33:1031 (1974).
2. J. M. Dale, L. D. Hulet, and S. Pendyala, Surf. & Interface Anal. 2:199 (1980).
3. I. J. Rosenberg, A. H. Weiss and K. F. Canter, Phys. Rev. Lett. 44:1139 (1980).

4. P. W. Zitzewitz, J. C. Van House, A. Rich, A. Koymen, D. W. Gidley, Physics Department, University of Michigan, Ann Arbor, private communication.
5. A. P. Mills, Bell Laboratories, private communication.
6. A. P. Mills, Proc. of the 83rd Sess. of the Intern. School of Phys. "Enrico Fermi", Varenna, Italy, July 14-17, 1981 (W. Brant and A. Dupasquier, eds.) to be published.
7. L. D. Hulett, J. M. Dale, S. Pendyala, Proc. of the Sixth International Conf. on Positron Annihilation, Fort Worth, TX, April 3-7, 1982 (P.G. Coleman, S. Sharma and L. M. Diana, eds.) North Holland, Amsterdam, The Netherlands.
8. S. Pendyala, Thesis (University of Western Ontario) University Microfilms Inc., Ann Arbor, Michigan, USA.
9. T. S. Stein, W. E. Kauppila, and L. O. Roellig, Rev. Sci. Instrum., 45:951 (1954).
10. J. K. Bair, P. D. Miller, and B. W. Wieland, Int. J. of Appl. Radia. Isot. 32:389 (1981)
11. L. D. Hulett, J. M. Dale, S. Pendyala, Surf. Interface Anal. 2:201 (1980).