INTRODUCTION

The existence of the positron was verified more than 40 years ago by Anderson but only in the 1950's was much attention given to its usefulness in terms of understanding the electronic structure of solids. As more workers became involved in the 1950's and 1960's work began to appear on metals, gasses and insulators and it was realized that positrons were sensitive to lattice defects. This realization has caused a new burst of interest in experimental and theoretical work using the positron as a probe for defects. Advantages of the positron technique are that it is nondestructive, very fast (10^6 counts in about 10 minutes), and highly sensitive to very low defect concentrations.

Initially the principal experimental techniques used were the measurement of the mean lifetime of positrons prior to their annihilation, and the angular correlation between γ-rays from two quantum annihilations. The use of Doppler energy broadening of the two gamma annihilation photons has emerged as a rapid experimental technique.

Many defects in crystals are known to have trapping effects on positrons. Vacancies trap positrons and this fact has been used to measure their formation energy. The missing positive ion results in an attractive potential for positrons. Vacancies and larger clusters are similarly expected to trap positrons and indeed current pulse annealing experiments following electron irradiation of copper single crystals in our laboratory is suggesting that these expectations were correct. Large annihilation rate changes have been reported due to volume defects, such as voids produced by neutron radiation of Al and Mo14,15,16. Dislocations produced by cold-work or fatigue are known to affect positron annihilation17,18,19,20,21,22. Grain boundaries contribute to positron trapping but stacking faults seemingly do not23. In what follows, I will describe very briefly the three main experimental measurement techniques used in positron annihilation and then describe some of the results from our laboratory. In this latter description the results quoted are mainly the efforts of former or current graduate students K. G. Lynn (fatigue of Ni and steel, grain boundary and fault work); D. Hadnagy (electron irradiation of Cu and voids in Al23); F. Alex (hydrogen embrittlement); P. Alexopoulos (copper fatigue) and of faculty associates R. W. Ure, Jr. and G. R. Miller.

* Paper read by Frank Alex of Hill Air Force Base, Ogden, Utah.
EXPERIMENTAL TECHNIQUES

Angular Correlation of Two-photon Annihilation Radiation

A typical set up for the measurement of the angular correlation of annihilation photons is shown in Fig. 1. A radioactive source $^{64}\text{Cu}$, $^{22}\text{Na}$, or $^{58}\text{Co}$ is placed close to the sample or perhaps the positrons are focused into the sample by a suitable magnetic field. The annihilation photons are detected by scintillation counters after passing through lead collimators to define the instrumental angular resolution. The coincident counting rate from the two detectors is measured as a function of the displacement of one detector.

If the annihilating pair is at rest in the center of mass system then the two quanta would be emitted precisely in opposite directions. We can assume for most purposes that the positron is essentially at rest after thermalization but the annihilation electron is not. This fact allows us to get at the momentum distribution of the electrons in the materials since the two photons will deviate slightly ($\sim 50$ mrad) from $180^\circ$. The detector is insensitive to the x and y momentum components but determines the z momentum component $P_z$. A knowledge of $P_z$ as a function of $\theta$ can be used to construct the Fermi surface for a material.

One example of the utility of this technique is the work of Berko and Plaskett$^{24}$ who were able to generate a theoretical angular distribution curve from calculated wave functions for: electrons in various shells of an atom, the valence electrons, and the positron. This curve for aluminum is shown as the solid line in Fig. 2 after normalization to the experimental points, which are the circles. The central parabolic part represents the angular distribution arising from annihilation with the three valence electrons of aluminum, while the outer curve represents the contribution from L-shell electrons. This illustrates the high momentum contribution to the angular distribution which comes from core annihilations.

The fact that the parabolic part is associated with lower energy electrons can be easily visualized by reference to annihilation in a free electron gas. We again consider the momentum of the photons to come from the electron since the positron is assumed to be at rest. The electron momenta are uniformly distributed throughout a sphere whose radius is the Fermi momentum. The conventional method of measuring an angular distribution corresponds to taking a slice through a sphere at a (transverse) momentum $K_z$. The area of this slice is shown in Fig. 3 and is proportional to the rate of annihilation into the region between $\theta$ and $\theta + d\theta$. But this area is proportional to $K_z^2 - K_0^2$, i.e. a parabolic dependence for a Fermi gas. The lower momenta then are associated with the parabolic part of the data and this part corresponds to the conduction or valence electrons.

The effect of damage to metals is to narrow the angular correlation curve i.e. cause more annihilations with lower conduction type electrons.
Fig. 1. Schematic diagram of a typical 2-photon angular correlation experiment.
Fig. 2. Angular distribution for aluminum. [111] data points are shown and the solid curve is a theoretical one normalized to the experimental counting rate at 0°. (After Berko and Plaskett).
Fig. 3. A slice through the Fermi sphere for a free electron gas.
because fewer ions, hence core electrons, are present in defect regions where the positrons tend to be trapped\textsuperscript{25,26}. It follows that by raising the temperature of a metal (and the vacancy concentration) the curve should narrow. This was first shown by MacKenzie et al in zinc, cadmium and indium\textsuperscript{27}.

**Doppler Broadening**

It was pointed out in the preceding section that the detectors used in angular correlation were sensitive only to $P_x$ or $K_z$, the component of electron momentum transverse to the direction of the emitted photons. In measuring along the direction of the emitted photons (x direction) one sees Doppler broadening of the 0.511 MeV annihilation photon depending on the momentum in the x direction of the electron at the time of annihilation. If the medium is isotropic with respect to the annihilation process (such as in a typical polycrystalline metal), then all momentum components are equivalent and $P_x$ would be equivalent to $P_z$ from angular correlation experiments. The most recent circuitry for Doppler broadening and considerable data on defects in metals are described in the Ph.D. thesis of P. Lichtenberger\textsuperscript{28}. A less sophisticated schematic equipment diagram is shown in Fig. 4 in which the detector is a lithium drifted germanium detector of high resolution. The best Ge(Li) detectors have resolutions of around 1 keV which is not nearly as good as that achieved with angular correlation. Nevertheless the Doppler technique has several advantages over angular correlation experiments. Only one detector is needed, no coincidence circuitry is necessary, data taking is ten times faster and weaker sources are possible than is the case in angular correlation.

It should be pointed out that in the case of damage to metals the Doppler peaks become sharper as more annihilations occur with conduction electrons in regions of defect traps which are lacking in positive ions. Figs. 5 and 6 illustrate the ideas of the Doppler measurement.

**Positron Lifetime**

Positron lifetime measurements involve a circuit such as seen in Fig. 7 in which the birth signal of the positron, 1.28 MeV, may be distinguished from the signal which accompanies its annihilation, 0.511 MeV, and the time interval between the two recorded. Since times as short as 2 or $3 \times 10^2$ picoseconds are often involved, an artificial delay of some nanoseconds is applied to the stop signal for convenience. Energy discriminators and a coincidence circuit lead to storage of valid pairs of events. A statistical distribution of these is accumulated over a period of about four hours (formerly 24) and the resulting curve is deconvoluted to remove the instrumental resolution characteristic. $^{60}$Co often is used to determine the resolution because it emits two time coincident $\gamma$ rays of approximately equal energy. The full width half maximum of the $^{60}$Co can be as narrow as 220 picoseconds. Slopes fitted to the exponential curve are reciprocal lifetimes as illustrated in Fig. 8 which shows some early raw fatigue data of Grosskreutz\textsuperscript{5}. Note that the fatigued copper
Fig. 4. Ge(Li) detector and circuit for Doppler-broadening lineshape studies.
I POSITRON ANNIHILATION IS ACCOMPANIED BY $\gamma$-RAY EMISSION

TYPICAL POSITRON SOURCE - Na$^{22}$

PRIOR TO ANNIHILATION

SOLID SPECIMEN

For a collection of annihilation events one measures the energy of the annihilation photons. This energy distribution will be Doppler broadened as a function of the momentum of the centers of mass of the electron-positron pairs.

Fig. 5. Schematic of positron annihilation.
II DEFECTS SHARPEN THE ENERGY DISTRIBUTION OF THE ANNIHILATION PHOTONS

![Diagram of positron annihilation in a material with and without defects](image)

As the content of defects, such as dislocations and vacancies, increases in a metal the defects serve as attractive trapping centers for positrons because the ion core density is lower in such locations. The Doppler broadened annihilation line shape will then narrow because (qualitatively) the defects allow a larger proportion of the positrons to annihilate with low-momentum conduction electrons which cause relatively little Doppler broadening.

Fig. 6. Schematic of positron annihilation in a material with dislocations.
Fig. 7. Schematic circuit diagram for the measurement of positron lifetime.
Positron decay curves in annealed and fatigued copper (99.1%).

Fig. 8. Fatigue data of Grosskreutz and Millett<sup>5</sup>. 
has a lesser slope, hence a longer lifetime than for the annealed Cu. It becomes the task of the researcher to extract and interpret these lifetimes in terms of trapping models of mechanisms occurring in the specimen. It should also be noted that very good temperature control is recommended to minimize electronic problems. We have utilized extra air conditioners and thermoelectric cooling of components to achieve this.

Figure 9 of West\textsuperscript{30} points out interesting differences in positron lifetime behavior between a typical polymer, an ionic solid and a metal. In polymers (also glassy solids, organic liquids and condensed gasses), lifetime spectra can usually be resolved into two or three components with lifetimes ranging from 0.1 to 10 nanoseconds. Ionic solids also show several components but individual ones are rather similar and are resolved with difficulty by the latest techniques. Metals usually have one or two lifetimes in the range 100-500 picoseconds, longer ones being ascribed variously to annihilations in the source salt or its mylar container. I would like now to present some samples of data obtained by the group at Utah to illustrate a number of possible NDE application areas.

RESULTS

Fatigue

Our earliest experiments involved plastic range fatigue cycling of nickel and nickel-cobalt alloys. The main effect is illustrated in Fig. 10 which shows, for a soft Ni-Co alloy, that a very early saturation effect occurred. This saturation is apparently one of the technique rather than a saturation in defects because Fig. 11\textsuperscript{21} shows that the X-ray particle size, which varies inversely with the dislocation density, does not saturate until about 75% of the fatigue life, whereas the positron lifetime essentially saturates in this case about 10% or less of the fatigue life. Thus for NDE purposes positron lifetime is rather conservative in that it responds perhaps too soon before failure. This is not the case, however, when a material is cycled within its elastic limit as was the case for 4340 steel\textsuperscript{30} initially 27 Rockwell C cycle at a maximum stress of 2/3 of its yield stress. Figure 12 shows that no saturation effect occurred and that, in fact, a final gentle linear increase in positron lifetime occurred which could be ideal for NDE estimates of remaining fatigue life for similar material.

When one cycles initially hard (R,51) 4340 steel\textsuperscript{30} a very interesting difference is found in that fatigue softening occurs producing a decrease in positron lifetime. Figure 13 shows a drop of some 40 psec and interestingly enough the previous curve and this one both reach 160 psec at fracture although from opposite directions.

In current experiments we are applying sub-yield stress fatigue cycling to polycrystalline copper with various degrees of cold work. Heavily cold rolled copper cycled at 1/2 of its yield stress showed only a slight decrease...
Fig. 9. Typical lifetime spectra. (a) Molecular materials. (b) Ionic solids. (c) Metals. Note the variation in time scales for these figures.
Fig. 10. Ni-Co fatigue data.\textsuperscript{21}

Fig. 11. Ni-Co x-ray particle size during fatigue.\textsuperscript{21}
Fig. 12. 4340 steel - lifetime response to fatigue hardening\textsuperscript{30}
Fig. 13. 4340 steel - lifetime response to fatigue softening$^{30}$. 

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Mean Life Time

vs Cycles of Fatigue

4340 Steel RC-51

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Mean Life Time $\tau \times 10^{12}$ Sec

Cycles $\times 10^{-3}$

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Fig. 13. 4340 steel - lifetime response to fatigue softening$^{30}$. 

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in positron lifetime over its total fatigue life of 8 million cycles. Evidently the relatively gentle fatigue cycling did not change the bulk dislocation density sufficiently out of the saturation range to permit the technique to respond well. We now are seeking to define more precisely the saturation region and to fatigue various hardness level specimens of copper.

**Hydrogen Embrittlement**

Various studies\(^1\) were conducted to determine if the effect of electrolytically induced hydrogen could be detected using the positron lifetime method.

Since the majority of classical hydrogen embrittlement tests have been made using commercial low alloy steels, A.I.S.I. 4340 steel was employed in our work. The positron lifetime variation with hardness or tensile strength was determined using specimens prepared by conventional quench and double temper heat treat to four hardnesses, including one which was fully annealed. Positron lifetime studies of these specimens gave the results in Table I. Embrittlement caused increases in positron lifetime at all hardness levels, probably as a result of stresses induced by hydrogen absorption.

Since dislocations and vacancies increase lifetime, the data noted are consistent with the fact that harder materials contain a greater defect density.

Next the effect of strain, hence dislocation density, was studied. This series of tests provided a most interesting result. In general the lifetime increased very rapidly with increasing strain reaching a maximum lifetime for all four hardnesses at approximately 3% to 7% elongation. Since the harder specimens either fractured or locally necked at approximately 7% elongation, they could not be studied beyond this point, the annealed specimen attained elongation of approximately 27% before fracture. As seen in Fig. 14, after reaching a maximum lifetime at about 3% elongation the curve then decreased until an elongation of 15% was attained, and then increased again. The decrease in lifetime was also exhibited by the Rockwell C 37 & 43 specimens but it's cause is not presently understood.

<table>
<thead>
<tr>
<th>Hardness</th>
<th>Before</th>
<th>After</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rockwell B 90.5</td>
<td>156</td>
<td>167</td>
</tr>
<tr>
<td>Rockwell C 37</td>
<td>168</td>
<td>180</td>
</tr>
<tr>
<td>Rockwell C 43</td>
<td>170</td>
<td>181</td>
</tr>
<tr>
<td>Rockwell C 51</td>
<td>175</td>
<td>180</td>
</tr>
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*Average of a varying number of tests over a two year period.
Fig. 14. Positron lifetime measurements at various tensile strains - 4340 steel.
It is interesting to note that the positron lifetimes of Table I peaked at about 181 picoseconds after charging. Some current literature concludes that hydrogen is trapped at dislocations. Hydrogen trapped as a positively charged ion would neutralize the negative charge associated with the core of a dislocation or vacancy. Since the negative charge is the reason a positron is trapped in these defects, this loss in charge would reduce the efficiency of dislocations as trapping sites depending on the concentration of hydrogen along the dislocation. The peaking noted in Table I may be a rather complicated combination of saturation of the technique at a given defect density modified by the unknown degree of the neutralizing effect of hydrogen on dislocation and vacancy traps.

**Electron Irradiation**

Electron irradiation\(^{13}\) of a copper single crystal containing an internal \(^{22}\)Na source have been performed in a 5 MeV beam. The crystal received a dose of 50 ppm Frenkel defects at 90\(^{0}\)K and was subsequently pulse annealed. This dosage produced saturation of the measurement. A subsequent 10 ppm radiation permitted a partial resolution of stage III structure during pulse annealing. The next irradiation will be even lighter to allow full development of the stage III structure.

**Voids in \(\alpha\) Alumina**

Lifetime measurements were performed on hot-pressed \(\alpha\) alumina specimens of varying density. Figure 15 shows a decrease\(^{13}\) of positron lifetime with increasing fraction porosity. This is quite the opposite behavior reported\(^{15}\) for neutron induced voids in metals in which case a lifetime increase is found. The reason is thought to be connected with the different surface state situation in \(\alpha\) alumina.

**Grain Boundaries**

The effect of plastic deformation on copper of varying grain size\(^{22}\) is shown in Fig. 16. Apparent saturation is being approached at fracture and the relative positions of the curves is consistent with higher work hardening in finest grain size materials. It should be noted that at zero strain the lifetime increases as one goes from single crystal to progressively finer grain size indicating the clear effect of grain boundaries on positron trapping.

**SUMMARY**

A number of examples of the considerable potential of positron annihilation for use in NDE have been shown. A possible limiting factor will be saturation effects of the phenomena at higher defect densities.

**ACKNOWLEDGEMENT**

The research quoted from the University of Utah is supported by ERDA(AEC).
Fig. 15. Positron lifetime measurements of various density α alumina specimens.
Fig. 16. Positron lifetime versus tensile strain for various grain size copper\textsuperscript{22}. 

Copper Samples 99.999\% Purity.
References

12. A. Seeger, Distinguished Lectures in Materials Science, ed. by L. Murr, and Ch. Stein, (Marcel Dekker, Inc. N.Y., N.Y., 1974).
18. P. Hautojärvi and P. Jauho, (to be published).


DISCUSSION

DR. TIEN (Henry Krumb School of Mines, Columbia University): Thank you. Any comments? I have one. John Wallace's Ph.D. thesis from Columbia showed there is a component called \( \tau_2 \) which is basically a longer lifetime/positron component which is very sensitive to dislocation and not so sensitive to point defects. Are you aware of all this?

MR. ALEX (Hill Air Force Base): Yes, we are aware of that, but we haven't been able to find a correlation between that and the positron lifetime due to annihilation in the metal.