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CRYOGENIC TEMPERATURE MEASUREMENT IN A NUCLEAR RADIATION ENVIRONMENT*

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ABSTRACT

A general discussion of the effect of radiation on the performance of platinum resistance thermometers, copper-constantan thermocouples and solid state devices is given. The radiation effects covered include rate phenomenon and integrated dose effects on the precision of the measurement, particularly in the cryogenic temperature range.

CRYOGENIC TEMPERATURE MEASUREMENT IN A NUCLEAR RADIATION ENVIRONMENT

INTRODUCTION

The more significant radiation effects will be discussed as they relate to influencing a temperature sensors performance in the cryogenic temperature range.

RADIATION EFFECTS IN GENERAL

Nuclear radiation primarily effects a transducer in two basic modes: 1. Those changes induced due to a prolonged exposure, such as material property changes; for example, mechanical properties, chemical properties, thermal and electrical properties. These changes are generally found due to "dose" effects; 2. Transient effects such as heating due to gamma or neutron flux rate, ionization, photo electric effects, induced charges in insulation are classed under "rate" effects.

Cryogenic temperature sensors most generally used are: 1. Resistance thermometers utilizing platinum or nickel; 2. Thermocouples such as copper-constantan, chromel P-constantan, normal silver-constantan; 3. Semiconductor resistive devices; carbon, germanium or junction devices.

Let us consider each of these three general approaches with respect to susceptibility and performance in the nuclear radiation environment.

RADIATION EFFECTS ON RESISTANCE THERMOMETERS

Platinum resistance thermometers constructed for severe environments such as vibration, shock, and wide temperature extremes are available and some of these units are termed by the manufacturers as radiation hardened. This is attempted by using only boron free ceramic insulation.

The selection of inorganic insulation such as Al_2O_3 , MgO or equivalent avoids

electrical insulation difficulties except for doses in excess of 10^{20} nvt. However, the sensing wire can exhibit effects below this dose level. The electrical resistivity of the platinum wire is effected by any disorder in the lattice structure since the electron wave is scattered by the perturbing non periodic potential. Neutron bombardment creates dislocations, knocking atoms out of the lattice leaving them in the structure elsewhere (Schottky and Frenkel defect) and at cryogenic temperatures, particularly below 20° K, the thermal vibration energy of the atoms is insufficient to restore the order. This restoration is termed "annealing effects." The % recovery of damage that was incurred at 4° K is plotted versus warming temperature or annealing temperature given by Blevitt, et al at ORNL is shown in Figure 1. It is seen that when platinum is raised to 24° K, 45% of the damage is annealed out.

T. H. Blevitt, R. R. Coltman and C. E. Klabunde^{9, 10, 11} at ORNL have irradiated platinum wire at 4.2° K for doses of 8×10^{16} nvt $E > 0.5$ MEV. The observed change in resistivity was linear with dose, amounting to 6×10^{-8} ohm-cm increase. Recent tests conducted at the Nuclear Aerospace Research Facility by Aerojet General on several platinum resistance thermometers at 20° K for the NERVA application¹² indicate also a linear resistivity change with dose, amounting to $8 - 10 \times 10^{-8}$ ohm-cm for approximately the equivalent dose as the ORNL data. This is shown in Figure 2. This is fair agreement considering the fact that the platinum tested in each case was from different sources and it verifies the expected of integrated accumulation of defects in the lattice structure. This phenomenon is not dependent upon the wire geometry but solely upon the material, its purity, lattice order, and the energy deposition from the incident radiation flux.

In addition, slow neutrons are captured and some atoms of proper isotope are transmuted. If the exposure is of sufficient length of time relative to the half life of the isotope and the following decay chain, an accumulation of impurity atoms will occur which will disturb the periodic potential and cause an increase in the resistivity.

C. W. Ross²³ tested thermocouple materials including platinum and platinum 10% rhodium in the material testing reactor to a flux of 4×10^{13} nv for approximately 70 days at a test temperature of 1000° F. Here the observed change in resistivity was due to transmutation, since at 1000° F all other defects are well annealed out. This is an excellent paper and I refer you to it in computing the change in resistivity due to transmutation.

Quite good agreement between test results and computation has been obtained.

Ross obtained for platinum:

$$\% \frac{\Delta \rho}{\rho_0} = 19 \times 10^{-22} \phi t$$

where: ϕ is the neutron flux in neutron/cm² sec, and t is the exposure time in seconds.

The above equation implies exposures times long with respect to the half lives of the isotopes of platinum, and the trace impurities initially in the wire.

W. E. Browning, Jr., and C. E. Miller, Jr.^{1, 2} of the Reactor Chemistry Division, Oak Ridge National Laboratory calculated the change in thermocouple composition for flux rates of 10^{14} nv up to 20 years of irradiation for 6 common thermocouples. The resultant change in resistivity due to this impurity concentration can be computed as shown by Ross²³ above. The computation includes the half lives of the isotopes generated. It was their general conclusion that platinum was relatively stable because of the number of platinum isotopes which are not radioactive. After a 10 year exposure, 1.85% of the platinum was converted to 0.32% Au and 0.92% Hg.

The platinum wire resistivity at LH_e or ρ_0 is very dependent upon its stress state and induced lattice dislocations due to "cold working" bending, etc. Manufacturers of precision platinum resistance thermometers anneal the wire at fairly high temperature after winding the sensing element to anneal out these defects. They also carefully select materials in the supporting structure and the ceramic insulator to avoid stressing the wire due to thermal coefficient differences. Radiation exposure of ceramic materials such as Al_2O_3 , BeO, etc. has resulted in some volumetric changes in the insulator. Although generally this is a very small effect, it should be considered when selecting ceramic cements and support structure so that these dimensional changes due to irradiation do not result in strain in the platinum wire.

Some decrease in the thermal conductivity of materials has been observed for materials under irradiation. In theory this is closely related to the electrical conductivity changes. The conductive heat transfer between the sensing element and the material to be measured may in certain critical situations be sufficiently altered to introduce an appreciable error.

The most significant rate effect is that of gamma and neutron energy deposition.

The gamma absorbed dose rate expressed in rads/hr or ergs/gm(c) is simply an

energy deposition referred to carbon. For other materials, the cross section correction must be made. The Kiwi and Nerva reactors utilizing LH_2 propellant present high γ fluxes for cryogenic temperature sensors, with resulting energy deposition in the order of 2 - 4 watts/gram (c).

Without a detailed description of the exact geometry of the sensor and the heat transfer conditions, an evaluation of the gamma heating error cannot be made. Computer calculations have been made on similar devices which include this distributed heat generation throughout the sensor and the heat flow paths available. Simple geometries can be treated by simple resistive lumped networks, or analog models can be constructed using a temperature voltage analog with electrical resistance paper.

The following is an example from recent test experience. A platinum resistance thermometer was used to measure the temperature of a large metallic member emersed in LH_2 . The platinum sensing element was housed in a 0.090" O.D. steel sheath of a few mills wall thickness. This sheath was placed in a milled slot and soft soldered with solder completely filling the gap between the sheath and the material providing excellent thermal contact between the two. Upon exposure to a γ flux of 10^{11} ergs/sec, however, the sensors read 150°R higher than the surrounding material. It was found that an air gap between the platinum wire and the outside sheath was sufficient to thermally isolate the wire and bobbin and the gamma induced heat was confined to the wire and produced a large temperature increase in the sensing element. No adequate solution to this problem has been found. This error could have been reduced to 30°R if the gap had been filled with H_2 or He. The difficulty lies in providing sufficient thermal conductivity between the sheath and the sensing element and yet not mechanically stressing the wire with the resultant resistance change due to stress, and yet adequately electrically insulating the wire.

Evaluation of various designs can be made by utilizing the self heating error as an indication of the effect of γ heating. The calculated γ heating can be approximately simulated by increasing the exciting current to the sensor and observing the indicated temperature increase while the thermometer is placed in a system duplicating the heat transfer conditions expected in the use environment.

The self heating can also be utilized to re-anneal the sensor; by applying a short pulse of increased exciting current, however, it is not practical where recalibration is not available in the system.

RADIATION EFFECTS ON THERMOCOUPLES

Thermocouples are simpler devices. The sensing junction can be made extremely small with the resultant improvement to minimize perturbation to the media, decreasing the response time, and the gamma and neutron heating can be reduced to a negligible effect in many cases. Measurements at Oak Ridge of copper-constantan thermocouples in Liquid He have shown negligible dose effect (less than 0.07°K error) for dose of 10^{18} nvt.

Despite these encouraging words, the old familiar disadvantages of thermocouples, i.e., thermoelectric power of only $3 \mu\text{V}/^\circ\text{K}$ at 10°K for copper-constantan and inhomogeneities in the thermocouple wire, make the use of thermocouples a very doubtful improvement for measuring cryogenic temperature.

Robert L. Powell⁴, et al., tabulated the maximum inhomogeneity emf as a function of wire length of #36 AWG wire for suitable cryogenic thermocouples.

In the 20°K range, #36 AWG constantan wire several inches long exhibited an emf of $0.9 \mu\text{V}$ and Cu wire $2 \mu\text{V}$. These would contribute a measurement error of $\approx 0.5^\circ \text{K}$.

R. J. Corruccini²¹ strongly recommends "normal silver" (0.37 at % Au) rather than copper due to the large increase in thermal conductivity of copper when decreasing temperature which may influence the lead conduction error. The thermoelectric power of copper is very sensitive to transition-metal impurities, and especially to iron at low temperatures. Very high purity copper has been seen to show variations of $6 \mu\text{volts}/^\circ\text{K}$ in its thermoelectric power at temperatures considerably below the Debye characteristic temperature where impurity atoms effect the electron wave scattering. At higher temperatures the thermal vibration of the atoms in the lattice dominate the scattering.

The calculations^{1, 2, 23} referred to in considering the radiation effects in platinum resistance thermometers, in particular that of transmutation of impurity atoms, also apply to the thermocouple. It has been shown in tests, however, that these effects are quite small. Some avoidance of spurious emf's can be made by choosing a suitable reference temperature, i.e., as close to the temperature to be measured as possible, and by not imposing large temperature gradients on the less homogeneous lead wires. These two suggestions reduce the Thompson emf's and spurious effects due to inhomogeneity and increase the precision of the measurement. Yet with extreme care it is doubtful that precision temperature measurement in the combined radiation plus cryogenic environment made with thermocouples can be better than $\pm 1^\circ \text{K}$. System noise is of major importance in assessing the practicability of the use of thermocouples. For example, a 1% accuracy at LH_2 with copper-constantan requires a system sensing capability of ± 1 microvolt.

Although this is quite practical in the Laboratory, it is a difficult achievement in the field and normal reactor instrumentation facilities.

RADIATION EFFECT ON SEMICONDUCTOR SENSORS

Semiconductor cryogenic temperature sensors have excellent sensitivity, particularly in the range of 100° R to LHe. Germanium crystals investigated by Powell, Cagwood, Bertram and Harden at NBS have demonstrated that these sensors have excellent thermal cycling stability variations of 0.002° in cycling to room temperature to 4° K and return. Carbon resistors in general have been found to have less reproducibility than Germanium. Unfortunately, Germanium has shown a high sensitivity to radiation induced resistivity and particularly at cryogenic temperatures.

There has been much work done in the study of radiation damage to Germanium and much work has been carried out at temperatures as low as 4.2° K. A large portion of this work has been done by investigators whose primary purpose was to determine the nature of damage and its mechanism rather than evaluate the effects of a combined environment. Electrical resistivity or conductivity is effected by both changes in carrier concentration and in the mobility. Cleland, Crawford and Pigg²⁷ irradiated n type Ge to 3×10^{16} n/cm² and they found that the carrier removal rate was higher by a factor of 1.5 for low temperature bombardment as compared to room temperature bombardment. They found that the rate of decrease of conductivity with exposure to fast neutrons at 16° K to be 3 times greater than the rate at room temperature.

Although there has been considerable work done in the last three years, a complete understanding of the complex mechanism of radiation damage of semiconductors has not yet been achieved. As a rough rule, if the dose exceeds 10^{15} nvt, this material is useless as a precise temperature sensor. For low dose exposures, it would be best to review the wealth of data now available prior to their use.

SUMMARY

Thermocouples are by far the least sensitive to radiation and unfortunately also to temperature. However, for an accuracy goal of +3° K, this is the simplest and most stable approach. Platinum resistance thermometers can be used in an attempt to increase the signal to noise ratio, and precision. However, considerable work in design and application must be expended to achieve improved performance.

Semiconductor sensors are of most interest in the temperature range below LN_2 where the sensitivity of the platinum resistance thermometer begins to falter. These are the most sensitive to radiation damage with a much lower threshold of damage. They are subject to all the problems associated with the platinum thermometer and more so. However, in certain applications their use may be warranted.

References

1. M. J. Kelly, W. W. Johnston, C. D. Baumann, "The Effects of Nuclear Radiation on Thermocouples," Oak Ridge National Laboratory, Oak Ridge, Tennessee.
2. W. E. Browning, Jr., C. E. Miller, Jr., "Calculated Radiation Induced Changes in Thermocouple Composition," Oak Ridge National Laboratory, Reactor Chemical Division, Oak Ridge, Tennessee.
3. D. S. Billington, "Radiation Effects in Metals and Alloys," Effects of Radiation on Materials edited by J. J. Harwood, H. H. Hausner, J. G. Moore, and W. G. Rauch, Reinhold Pub. Corp., New York, 1958, Chapter 3.
4. R. L. Powell, Lindsay P. Cagwood, Jr., M. D. Bunch, "Low-Temperature Thermocouples," Cryogenic Engineering Laboratory, NBS, Boulder, Colorado.
5. G. F. Levy, R. R. Fouse, R. Sherwin, "Operation of Thermocouples Under Conditions of High Temperature and Nuclear Radiation," Marquart Corporation, Van Nuys, California.
6. F. D. Werner, "Some Recent Developments in Applied Platinum Resistance Thermometry," Rosemont Engineering Company, Minneapolis, Minn.
7. G. T. Furkawa, "Application of Resistance Thermometers to Calorimetry," NBS, Washington, D. C.
8. H. B. Sachse, "Measurement of Low Temperatures with Thermistors," Keystone Carbon Co., St. Marys, Pa.
9. P. J. Bendt, "Liquid Hydrogen Thermometers in a Radiation Field," Los Alamos Scientific Laboratory memo, CMF-9, dated 6 Nov. 1961, P. J. Bendt to E. F. Hammel.
10. Letter dated Feb. 22 1961, T. H. Blewitt, consultant Oak Ridge National Laboratory, to Dr. A. M. Liebschutz, NASA Cryogenic Project Manager, Lockheed Nuclear Products, Dawsonville, Georgia.
11. T. H. Blewitt, R. R. Coltman, C. E. Klabunde, "Solid State Div. Ann. Progress Report for Period Ending Aug. 31, 1958, ORNL-2829 and ORNL-2614, Oak Ridge National Laboratory, Oak Ridge, Tennessee.
12. NERVA Components Irradiation Program, Volume 7: GTR Test 9, FZK-170-7, 30 Dec 1963, prepared by Nuclear Aerospace Research Facility General Dynamics Fort Worth, Test No. 23/W105, Resistance Thermometer tests.
13. E. R. Nolte, H. C. Chandon, "Evaluation of Rosemont Engineering Co., Model 134EB Radiation Resistant Cryogenic Temperature Sensors," Laboratory Report 8771:2365, Jan. 8, 1964, Aerojet General Corporation Liquid Rocket Plant, Sacramento, California.

14. V. A. J. VanLint, "Mechanisms of Transient Radiation Effects," prepared for the Summer General Meeting of the IEEE, Toronto, Canada, June 17-21, 1963, General Atomic div of General Dynamics GA-4320, Paper no. 63-1180.
15. N. F. Mott, H. Jones, "The Theory of the Properties of Metal and Alloys."
16. "NERVA Components Irradiation Program," Vol. 1, GTR Test 4, FZK-170-1, July 19, 1963, prepared by Nuclear Aerospace Research Facility, General Dynamics, Fort Worth.
17. G. L. Fluke, "Winsco Platinum Resistance Temperature Transducer Model 2610-2," Report no. 8771:2256, Dec. 17, 1963, Aerojet General Corporation, Liquid Rocket Plant, Sacramento, California.
18. Battelle Memorial Institute, REIC-2, "The Effect of Nuclear Radiation on Structural Materials," September 1961.
19. R. Bowman, "The Effect of High Level Radiation on Instrumentation Transducers," Wiancko Engineering Co., presented at Aerospace Instrumentation Seminar, Lockheed Missiles and Space Corporation, May 1, 1963.
20. R. E. Bowman, "How Radiation Affects Engineering Materials," REIC Battelle Memorial Institute, Materials in Design Engineering, July, 1960, Reinhold Publishing Co.
21. R. J. Corruccini, "Temperature Measurement in Cryogenic Engineering," C. E. L. N. B. S., Boulder, Colorado.
22. P. J. McNally, "The Effect of Nuclear Radiation at Cryogenic Temperature on Semiconductor Materials," REIC No. 30, September 30, 1963, Battelle Memorial Institute.
23. C. W. Ross, "Effect of Thermal Neutron Irradiation on Thermocouples and Resistance Thermometers," Leeds and Northrop Co., Philadelphia, Pa., AIEE, paper No. 5-4, presented at the Joint Nuclear Instrumentation Symposium, Raleigh, N. C., Sept. 8, 1961.
24. P. H. Hunter, "Resistance and Temperature Measurement," North Carolina Works, Electrical Standards Laboratory.
25. O. Sisman, C. D. Bopp, R. L. Towns, "Radiation Stability of Ceramic Materials," ORNL Solid State Progress Report for period ending Aug. 31, 1957.
26. J. S. Blackmore, J. W. Schultz, J. G. Myers, "Review of Scientific Instruments," Vol. 33, 545 (1962).
27. J. C. Figg, "Annealing of Radiation Effects on Germanium," Oak Ridge National Laboratory, Oak Ridge, Tennessee, ORNL-2829, Solid State Div., Annual Progress Report, Aug 31, 1959.

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FIGURE 2. RELATIONSHIP BETWEEN GAMMA DOSE AND CALCULATED DOSE
TEMPERATURE DEVIATION VS. GAMMA DOSE

FIGURE 4. 7-MILLOE VOLTAGE

VS. TIME