RADIOCONDUCTIVITY OF INSULATORS:
Their Potential for Ionographic Imaging

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Abstract Measurements of the radioconductivity and thermocurrent properties of relatively pure insulating solids indicate charge collection yields which compare favorably to materials presently used for ionography. The radioconductivity varies with temperature giving rise to temperature windows of feasible use.

These experiments were unanticipated in the last grant proposal. As a result of parallel work supported by other resources, the potential for x-ray imaging was discovered and explored. The implication for practical applications, however, were immediately obvious in their relevance to this program.

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Introduction

When any dielectric material, solid, liquid, or gas, is exposed to ionizing radiation, mobile electrical charges result. In an applied electric field, these charges drift to produce an electrical current; this process can be termed radioconductivity (RC). In gases, this phenomenon is the basis of air ionization chambers; in highly purified semiconductor solids, it provides an extremely sensitive method for detecting even single event x or Y-ray absorptions.

This report describes preliminary radioconductivity studies for some high purity insulating solids. One baseline for comparison is given by the radioconductivities of the nominally pure solids which were available several years ago. Huntley and Andrews¹ review many experiments and show that charge collection yields were on the order of 10^{-16} coulombs/cm²-mR for a standard applied electric field of 500 volts per millimeter. This value was typical of a variety of insulating solids and was not very dependent upon the temperature of the solid samples.

Another baseline for comparison is the ionographic charge yield for the liquid insulator tetra-methyl-tin (TMT), which Fenster and Johns² evaluated as an excellent radiographic imaging system. For 4 mm of TMT, and an applied field of 10^4 volts/cm, they measured a charge yield of 10^{-9} coulombs/cm²-mR. Of course, the fields applied to the liquid are 20 times greater than those used to measure solids, but even with this taken into account, the charge yields characterizing the liquid state radioconductivity are still 10^5 - 10^6 larger than those which characterized solids. Our early experiments on older solid samples gave radioconductive behavior in reasonable agreement with that

described above. It is for this reason that high atomic number, ionic solids have not been heretofore considered as potential ionographic detectors. However, our recent experiments have shown that this small sensitivity is not characteristic of the solids but is rather the result of having large concentrations of certain kinds of impurities.

Recently much purer solids than those of the past have become available, and their thermocurrent (TC) and RC properties are much greater. Our recent measurements on selectively purified insulating solids give charge yields a factor of 10^6 greater than previously observed; We actually have observed, in ordinary (but high purity) ionic insulators, charge yields rivaling those obtained in ultra high purity semiconductors. We wish to describe these studies and their implications for imaging.

Background

It is widely believed that these previously observed small currents occur because the electron-hole pairs (the mobile charge entities in a solid) can rapidly trap into immobile states at impurity or defect centers and because recombination of the positive-negative charge pairs can occur so rapidly in a condensed medium. Thus, when irradiation ceases, the solid retains some charges trapped in immobile states, but these charges are only metastably trapped. By heating the material, these metastably trapped charges become thermally remobilized, producing transient electrical currents flowing at a number of characteristic temperatures related to the trapping depths of the metastable traps. These remobilized currents give rise to thermocurrent peaks (TC) and they can be read out and utilized as a radiation dosimetry technique.³⁻⁵ Remobilized charges drift until they retrap at even deeper

trapping or recombination centers. This process limits the radiation sensitivity of the TC effect. If the deep traps and/or recombination centers are optically active, then light emission results at the end of the thermally remobilized lifetime. This is called thermoluminescence (TL) and is the presently most practically successful of the solid state passive radiation dosimetry techniques.⁶ Thermoluminescence dosimeters are doped intentionally with high concentrations of activator impurities to give fast and efficient, optically active recombination and trapping.⁷

Just as for the radioconductivity signals during irradiation, until very recently the thermocurrent signals read out subsequent to irradiation had been very weak in relation to gaseous state or semiconductor ionization currents.³ For comparison, a one Roentgen exposure for an air chamber into a collection volume of 0.1 cm^3 yields a charge collection of approximately 10^{-11} coulombs.⁸ On the other hand, one would typically collect less than 10^{-13} coulombs in a nominally pure crystal of Al_2O_3 , 1 mm thick by 1 cm^2 in cross-section (also 0.1 cm^3 in volume).^{1,8}

Results and Discussion

The TC/RC reader that was used for these experiments is similar to that described previously in report C00-1105-229.⁹ The x-ray tube potential was 80 kVp (filtered to give 35 KeV effective photon energy) with an exposure rate of approximately 300 mR sec^{-1} . The electrode had a collection area of about 0.33 cm^2 and a field of 400 volts/mm was applied.

Adolph-Meller U-V grade Al_2O_3 was irradiated and the thermocurrent peaks were measured (Fig. 1); note the presence of a signal at room temperature. These TC peaks correspond to temperature regimes where trapped charge is

thermally remobilized. In these temperature regions, the radioconductivity of the samples can become very very large. Figure 2 illustrates the equilibrium current which flows while the sample is exposed to x-rays at various temperatures in the region of the lower temperature peak of Figure 1. The difference between the equilibrium and maximum current, shown in Figure 2, results from a polarization in the sample.¹⁰ Subsequent to the application of an electric field and radiation, a constant current will flow after a given time period, which varies with the temperature during irradiation; this current is called the equilibrium current. However, if the sample is irradiated for this same time period and then the electric field is applied, the current is actually greater than this equilibrium value. This maximum current decreases with time, as the sample polarizes, to approach the equilibrium current. Note from Figure 2 that both of these currents exhibit the same temperature characteristics. These measurements were done for various electric fields and the currents had a linear relationship with the electric field for all temperatures.

Note from Figure 2 that the charge yield rises with increasing temperature to give a broad working "window" in the temperature scale. Similar studies have been done for CaF_2 where the TC peaks occur at about 90 K.¹⁰ However, in the case of CaF_2 , in the region of liquid nitrogen temperatures, as the temperature is increased, the charge yield decreased due to thermally stimulated retrapping of charges in a deeper set of levels. Consequently, there are temperature "windows" within which the solid shows practically useful radioconductivity. At too high a temperature the charge yield starts to be more limited by retrapping into deeper traps. Thus, the size of the temperature

window is important. Note that for Al_2O_3 , there is a broad window in the temperature scale.

As can be noted from these figures, the charge yield appears to be very favorable. The charge yield for Al_2O_3 is about 10^{-10} coulomb/cm²-mR for an applied field of 500 volts/mm. This is a factor of 10 larger than for the CaF_2 samples which have a value of 10^{-11} coulombs/cm²-mR at liquid nitrogen.¹⁰ This charge yield for Al_2O_3 is very competitive with the TMT liquid system yields. Note that these charge yields are 10^5 to 10^6 larger than that typical for nominally pure samples mentioned earlier. This result shows that, in principle with presently available solid state purities, ordinary insulating solids will perform at least as well as the liquid state systems for radiographic imaging.

We feel that these high conductivities can be explained by the following reasoning, which our experiments tend to support. The impurity trapping states effectively govern the TC and radioconductivity charge transport. In any particular solid, at a particular temperature, there will be a series of low binding energy traps which are thermally unstable, and a remaining group of deeper energy traps which are thermally stable. These thermally unstable traps may modify the time dependence of radioconductivity response, but do not limit the observed final sensitivity. Only the deeper stable traps, which cannot thermally release trapped charges, suppress the charge yield measured over some reasonable time. However, we have found that the trapping cross-sections of the deep, stable traps are extremely (and non-linearly) dependent upon the concentration of the particular impurity involved and sometimes upon temperature. This is a very important factor. The theoretical reasons are complicated and haven't fully been explored, but the empirical

results are clear. If impurity levels have concentrations typically greater than a few hundred parts per million (ppm) the radioconductivity yield is very small and not very dependent on concentration. With materials presently becoming available with active impurity concentrations of a few tens of ppm, the radioconductivity yield abruptly increases by a factor of 10^4 - 10^6 .

The key fact here is that the ionic material need not be of semiconductor purity quality. Even the modest improvements in purity available in the last few years abruptly switch typical ionic solids from the essentially useless radioconductive sensitivity measured in the past, to the high and potentially very useful sensitivity we recently have measured. Moreover, measurements of the thermocurrent effect combined with radioconductivity measurements allow us to determine the full characteristics of the trapping levels in a given solid. This gives us the possibility of specifying which levels are important for determining sensitivity, and which are not.

Ionographic Images

Although the Al_2O_3 effective atomic number is very small for efficiently absorbing x-rays of diagnostic energies, these results prove the feasibility of room temperature operation of solid state ionographic systems. The performance, for example, of the Al_2O_3 solid, operated at 5 kV across a 1 mm detector thickness, will be very similar to the TMT system evaluated by Fenster and Johns.¹²

A theoretical estimate of the probable performance of a solid in comparison to other ionographic materials can be made based upon the photon flux in an x-ray spectrum and the absorption characteristics of the ionographic material.

The relevant equations are given by Johns, et. al.¹¹ The two quantities of interest are the efficiency of absorption η , and the efficiency of picture taking, η_p . The value of η_p is the important quantity for ionographic images and includes the effect of the K fluorescent radiation which can "fog" the image. Materials having lower effective atomic numbers should not be excluded from consideration, since they have lower fluorescent yields and the radiation is all locally absorbed to aid in producing the image.

The calculated value of η_p for the Al_2O_3 sample of approximately 1 mm thickness was about 15%. The value given by Fenster and Johns for the TMT liquid for η_p is 40%;¹² however, this is for a 1 cm thickness of liquid. The values for high pressure gases are comparable.¹³ If the same thickness is used for Al_2O_3 , i.e. 1 cm, the picture taking efficiency is approximately 65%. This fact and the favorable charge yield of Al_2O_3 prompted us to attempt to make an ionographic image. Fig. 3A shows the first image of a ledex strip on a 1.5 cm diameter disc of Al_2O_3 ; Fig. 3B is a later image of a JEDEC -JT-4.2 resolution pattern made with Al_2O_3 a 2.5 cm x 2.5 cm Al_2O_3 plate obtained from Tyco Company. This is a rough image with artefacts present, but we feel they can be eliminated by the use of a transfer process. This is described in more detail in another publication.¹⁴

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Figure 1: Thermocurrent peaks in high purity Al_2O_3 . Note the presence of a signal at room temperature.

Figure 2: Al_2O_3 radioconductivity versus temperature for an applied field of about 400 V/mm.

Figure 3A: Ionographic image of a ledex strip using a 1.5 cm diameter disc of Al_2O_3 .

Figure 3B: Ionographic image of a JEDEC-JT-4.2 resolution pattern on a 2.5 cm X 2.5 cm plate of Al_2O_3 . The smallest bars are about 4 line pairs/mm.