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THE RADIOELECTRIC EFFECT (Information Report)



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THE RADIOELECTRIC EFFECT

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ABSTRACT

An electrostatic field will exist between two materials having different work functions when the only connection between the materials is through a load impedance. If the two materials are connected externally by a current-measuring device and the separating medium is ionized by exposure to radiation, a current of electrons will flow in the external circuit from the material of lower work function to the material of higher work function. The quantity of this current depends upon the value of the external load impedance, the type and intensity of the incident radiation, the difference between the work functions of the electrode surfaces, the type and pressure of the separating gas, and the geometry of the setup.

Radiation-measuring devices called "radioelectric cells" have been constructed on the basis of this "radioelectric effect". Gamma detecting cells have been developed which for specific uses offer marked advantages over conventional detectors. It is also possible to detect alpha, beta, and neutron radiation and to determine the difference between the work functions of two surfaces.

INTRODUCTION

An electrostatic field will exist between two materials having different work functions when the only connection between the materials is through a load impedance.^{1,2} It is well known that the magnitude of this field will be determined by the magnitude of the difference between the two work functions and by the distance between the two materials. Electrostatically, the material of lower work function will be positive with respect to the material of higher function because of the greater ease of removal of electrons from the former. If the two materials are connected externally by a current measuring device, no steady current flow is possible since electrons cannot flow through the separating medium. However, Kelvin showed that, if the separating medium is forcibly ionized by exposure to radiation, a current of electrons will flow in the external circuit from the material of lower work function to the material of higher work function. The positive ions formed in the separating medium drift toward and are collected by the material of higher work function; while the negative ions will tend to drift toward and be collected by the material of lower work function. As positive ions are collected they will be neutralized and, hence, will remove electrons from the material of higher work function. Those electrons lost in neutralizing the positive ions will be replaced by electrons flowing in the external circuit from the material of lower work function to the material of higher work function. Conservation of charge is maintained by the collection of the negative ions by the material of lower work function. This discussion ignores the small charge contribution resulting from possible differences in the numbers of electrons directly emitted by the electrodes through interaction with the ionizing radiation and any charge introduced in the system when the ionizing radiation consists of charged particles.

We have called this phenomenon of current flow the "radioelectric effect" and the complete device, exclusive of the external circuit, a "radioelectric cell".

The quantity of current which will flow in the external circuit has been found to depend upon the value of the external load impedance, the type and intensity of the radiation incident upon the cell, the difference between the work functions of the two electrode materials, the type and pressure of the filling gas, and the geometry of the cell. If the cell is excited by gamma radiation, the current is also a function of the ability of the electrodes and of the filling gas to produce secondary electrons. With neutron radiation, the current depends upon the ability of the electrodes and of the filling gas to produce ionizing particles by interaction with neutrons.³

Although the various factors which affect the magnitude of the current in the exterinal load suggest many possible uses for this effect, the prime effort at Mound Laboratory thas been directed toward determining the design factors necessary for the development of useful, practical, radiation-measuring devices. Gamma-sensitive instruments which are simple, efficient, and have high geometry factors have been developed. Several instruments are in routine use at Mound Laboratory and offer definite advantages over conventional instruments. Considerable data have been assembled on the design considerations of cells for neutron detection, but much further work will have to be done. With alpha radiation as the exciting source, currents as high as 4 microamperes through a load of 2,000 ohms have been achieved.

BASIC EXPERIMENTAL DATA⁵

Most of the measurements of cell characteristics were made with the cells under test connected as shown in Figure 1. In the early measurements a Beckman Ultrohmeter was used as the indicating electrometer. Later measurements were made with a Leeds and Northrup developmental electrometer (on field test), with a vibrating reed electrometer, and more recently with a newly-designed direct-current amplifier⁸ which has approximately 90 per cent negative feed back.

With the cell connected as shown in Figure 1, two important characteristics can be measured. With no voltage applied from the potentiometer and the cell exposed to radiation, a current flows through the cell and through the external load resistor, causing a voltage to be developed across this resistor. This voltage is called the "developed voltage".

The second characteristic is measured by adjusting the polarity and magnitude of the external voltage source until the current through the irradiated cell is reduced to zero, as indicated by the developed voltage dropping to zero. This bucking voltage is then equal in magnitude, but opposite in sign, to the open-circuit voltage of the cell. This opencircuit voltage is called the "characteristic potential" of the cell, since it is determined by the composition of the electrodes and the nature of their surfaces, but is independent of all other factors except for the possible presence of a third type of surface which can collect ions and produce circulating currents. This last phenomenon has been called the "third-electrode effect", and it usually results in a lower value of characteristic potential for a cell than would be obtained in its absence.

The developed voltage, on the other hand, is dependent on a large number of conditions. It varies with the type and intensity of the radiation, the characteristic potential and the geometry of the cell (including the number, spacing, and thickness of the electrodes), the nature and pressure of the filling gas, and the value of the external load impedance. With gamma radiation, the developed voltage also varies with the absorption coefficient of the electrodes and of the filling gas. With neutron radiation, the developed voltage also varies with the ability of the electrodes and of the filling gas to produce ionizing radiation upon neutron bombardment.

Early measurements of cell characteristics were made with the open-type cell shown in Figure 2; for clarity, the center electrode has been removed from the center of the cell and placed beside the outer electrode. The outer electrode (at ground potential) was formed in the shape of an open box, 3 by $7\frac{1}{2}$ by $7\frac{1}{2}$ inches, while the center electrode consisted of a flat plate, $4\frac{1}{2}$ by 5 inches (usually $\frac{1}{32}$ inch thick), supported in the center of the outerelectrode box by a Teflon insulating pillar. The inner electrode was connected to the electrometer through a shielded cable with insulation of high resistance. Two types of outer







electrodes.were.used. The first type was of lead which had been electrolytically cleaned. This electrode did not appear to be as stable with time as might be desired. The second was made of 2-S aluminum sheet and was given no special treatment. It appeared to be quite stable in its characteristics.

At first the top of the cell was left open, but it was found that air currents in the "laboratory could result in erratic readings. Later measurements were made with the top of the cell covered, usually with the same material as the outer electrode but sometimes with an insulating material.

Center electrodes were made in several ways. The electrodes were cut from a sheet of the desired material when stock was available. Otherwise, the desired material was plated on a brass base, except for the lead dioxide electrodes which were made by plating gold on a copper base and then plating lead dioxide on the gold by anodic deposition from a lead nitrate bath.

A silver-110 gamma source (with a tolerance of about one inch for eight hours) was used for most measurements because it was convenient, even though the cell.will.detect alpha, beta, and neutron radiation as well. The source was placed on the outside of the open cell at the bottom center of one of the long sides, when aluminum was used as the outer electrode. When lead was used as the outer electrode, the source was suspended inside the cell at the center of one of the long sides, to avoid absorption of radiation by the lead.

The measured characteristic potentials of an open-type cell with an aluminum (aluminum oxide) outer electrode, air filling, and with various center electrodes are shown in Table I. It will be noted that the various materials tend to fall in the order that would be predicted from the values for the work functions of the corresponding elements, though there are exceptions (see nickel and chromium). Such exceptions would be expected if the characteristic.potentials are a measure of the difference in the work functions of the surfaces of the electrodes, since in most cases these surfaces were oxides (or other compounds) rather than the pure element itself. Thus, two electrodes made of the same material, but having a different history, might have different work functions for the two surfaces and, hence, would show a small characteristic potential if used in a radioelectric cell. Such a condition has been found to be the case, and several instances are listed in Table I. Note, for example, oxidized copper, copper, and brushed copper. If the surface film is very thin, there may be a type of third-electrode effect, and the characteristic potential may have a value intermediate between the one it would have with a "thick" surface film and the one it would have with the pure element. Thus, two electrodes cut from the same sheet of aluminum will show essentially.a.zero characteristic potential; but if one electrode is abraded and the characteristic potential measured as rapidly as possible, a significant characteristic potential will be found. This potential decreases rapidly at first, then more slowly (approximately logarithmically), and eventually becomes essentially zero once more, as the oxide film again becomes sufficiently thick.

The characteristic potential of a specific radioelectric cell should be independent of the type of radiation and of the flux density. To show this independence, a lead dioxidealuminum (oxide) cell was set up and energized with gamma, recoil-proton, and alpha radiation. With gammas from the silver-110 source, the characteristic potential was 1.18 volts. With recoil protons (obtained by covering the top of the cell with a sheet of Lucite and bombarding it with neutrons), the characteristic potential was 1.17 volts. After minimizing third-electrode effects by covering the window of a polonium alpha source with masking

tape, except for a small hole to allow the alpha particles to pass through, the source was placed inside the cell and the characteristic potential was found to be 1.16 volts. Since these sources varied widely in flux density, this test also indicated that the characteristic potential is independent of flux density as well as the type of radiation.

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TABLE |

CHARACTERISTIC POTENTIAL OF CELLS WITH ALUMINUM (OXIDE) NEGATIVE ELECTRODES

| | WORK FUNCTION | | |
|--------------------------------------|--|--|--|
| TEST ELECTRODE | WORK FUNCT!ON ⁷ (volts) | REFERRED TO Aluminum As Zero (volts) | CHARACTERISTIC POTENTIAL (volts) |
| LEAD DIOXIDE PLATED ON GOLD | - | • | *1.34 |
| CARBON (AQUADAG COATED ALUMINUM) | 4.39 | 0.65 | 1.01 |
| COPPER OXIDE ON COPPER | • | • | 0.975 |
| GOLD PLATED ON COPPER | 4.58 | 0.84 | 0.95 |
| BRUSHED GOLD PLATED ON COPPER | - | 0.84 | 0.945 |
| COPPER | 4.47 | 0.73 | 0.72 |
| SILVER | 4.28 | 0.54 | 0.67 |
| BRUSHED COPPER | * | 0.73 | 0.64 |
| NICKEL | 4.84 | 1.4 | 0.63 |
| BRASS | . • | • | 0.58 |
| STAINLESS STEEL | o | • | 0.57 |
| BRUSHED BRASS | • | | 0,425 |
| SANDED NICKEL | • | 1.1 | 0.41 |
| NICKEL, ONE PER CENT COBALT (PLATED) | - | 1.4 | 0.41 |
| SANDED STAINLESS STEEL | - | ø | 0.39 |
| ZINC | 3.74 | 0.0 | 0.82 |
| LEAD | 4.02 | 0.28 | 0.22 |
| BRUSHED LEAD | ø | 0.28 | 0.20 |
| CHROMIUM (PLATED) | 4.51 | 0.77 | 0.18 |
| CADMIUM (PLATED) | - | 0.18 | 0.132 |
| CADMIUM | 3.92 | 0.18 | 0.06 |
| SANDED CADMIUM | • | 0.18 | 0.026 |
| SANDED ZINC | • | 0.0 | .∝ 0. €1 |
| MAGNESIUM | 3.46 | 0.28 | -0.26 |
| ALUMINUM (ONE MINUTE AFTER SANDING) | 3.74 | 0.00 | - -0.32 |

As a further check on the effect of flux density on characteristic potential, the cell shown in Figure 3 was investigated. This first multiple-electrode cell (called Model 1) consisted of interleaved gold and lead electrodes in a lead case. The gold electrodes were made by plating gold on brass, and the lead electrodes and outer case were made by plating lead on brass. The characteristic potential with the silver-110 gamma source was 0.45 volt. When the cell was energized by a 24.8-milligram radium source, this voltage again measured 0.45 volt.

The developed voltage of a cell with 0.82-volt characteristic potential was measured with several values of load resistance as shown in Figure 4. It can be seen that, as the load resistance was increased, the developed voltage approached the characteristic potential asymptotically.

To determine the effect of gas type and pressure on the sensitivity and the characteristic potential of a cell, a lead dioxide-aluminum (oxide), open-type cell was placed in a vacuum chamber so that the gas pressure could be reduced and the operation of the cell in dry air and argon atmospheres could be examined. When the initial measurements were made at atmospheric pressure (using the silver-110 gamma source), the characteristic potential of this cell was 1.28 volts and the developed voltage across 1.06×10^{11} ohms was 112 millivolts. When the chamber was pumped down to the limit of a Pressovac, the developed voltage dropped to less than one per cent of the original value and the characteristic potential could not be measured. When the chamber was filled with air dried through a calcium chloride tube, the characteristic potential was again 1.28 volts; but the developed voltage had increased to 126 millivolts. In an argon atmosphere, the characteristic potential was still 1.28 volts, but the developed voltage was 178 millivolts.

Some experiments were run to see if the cells were temperature sensitive by testing an open-type cell at room temperature and at 100° C. No change in characteristic potential could be detected within the precision of our measurements.

The lead dioxide-lead, open-type cell was connected directly to a galvanometer that had a sensitivity of 6.6×10^{-12} ampere per division and exposed to the 24.8-milligram radium source to see if a current strong enough to be measured directly could be produced. When connected in one polarity, a current of 1.3×10^{-10} ampere was observed and when connected in the opposite polarity, a current of -1.25×10^{-10} ampere was observed. When the Model-1, multiple-electrode cell was tested under the same conditions, currents of 1.45×10^{-10} and -1.39×10^{-50} ampere were observed. This change in cells showed a sizable change in the current-to-volume ratio, since the multiple electrode cell has only about $\frac{1}{20}$ the volume of the open cell.

The sensitivity of a cell to polonium alpha radiation was checked by placing alpha standards within a lead dioxide-aluminum (oxide), open-type cell and observing the developed voltage across 10^{11} ohms of resistance. The following deflections were noted: a sample giving off 100,000 particles per minute gave a voltage of 145 ± 5 millivolts, 10,000 per minute gave 35 ± 3 millivolts, 3,000 per minute gave 18 ± 1 millivolts, and 1,190 per minute gave 5 ± 1 millivolts.

With the lead dioxide-aluminum (oxide) open-type cell, the beta standards which we had did not produce sufficient current to be observed. The interleaved, flat-plate-type cell described later (Figures 5 and 6) was, therefore, used for the measurements. The electrodes were lead dioxide and aluminum (oxide), and the filling gas was air at atmospheric pressure. The following readings were made across 10^{11} ohms: a carbon-14 sample





MODEL 1 CELL, DISASSEMBLED





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INTERLEAVED FLAT PLATE CELL, DISASSEMBLED

of about 500,000 disintegrations per minute gave a reading of 12 millivolts, and a sulfur-35 sample of about 180,000 disintegrations per minute gave a reading of 8 millivolts.

A cell was tested for neutron counting and differentiation against gamma excitation by setting up a lead dioxide-lead, open-type cell with a neutron source suspended above it. An aluminum plate was placed between the cell and the source so that any recoil protons from the plastic case on the source would be blocked from the cell, but the gamma radiation from the source would still pass through the cell. Under these conditions, the developed voltage across 10^{10} ohms was 38 ± 2 millivolts. When a sheet of Lucite was placed between the aluminum plate and the cell so that recoil protons would enter the cell, the developed voltage was 53 ± 1 millivolts. A difference of 15 millivolts was thus caused by recoil protons. In an attempt to make the cell less sensitive to gamma radiation and thus to increase the differential caused by recoil protons, the above experiment was repeated with a copper oxide-aluminum (oxide), open-type cell. In this case, the gamma radiation alone gave a developed voltage of 26 ± 4 millivolts, and when the Lucite was added the developed voltage was 76 \pm 2 millivolts, a differential of 50 millivolts. This experiment illustrated how the gamma stopping power of the electrodes affected the sensitivity of a cell for gamma but should not for other types of radiation. This peculiarity with gamma radiation is caused because the gas in the cell is also ionized by secondary electrons emitted by the electrodes when struck by gammas, while in the case of alpha, beta, or neutron (by means of recoil proton) detection, the particle itself is the only ionizing agent.

Since it was established that increased current could be obtained by placing electrodes in parallel, the gamma-sensitive cell shown in Figure 7 was assembled to see if the characteristic potential could be increased by placing electrodes in series. When excited by the 24.8-milligram radium source, the open-circuit voltage, which had been 0.95 volt for a single, lead dioxide-lead, air-filled cell, was 1.62 volts. This value is considerably below what would be expected for five cells in series, and may have resulted from circulating currents within the cell since the polyethylene insulating sheath did not completely cover the lead. The fact that the cell was in a very nonuniform flux may also have been a contributing factor.

DESIGN CONSIDERATIONS FOR PRACTICAL CELLS

To determine the effect of the characteristic potential of a cell on the current flow (hence, developed voltage), an external source of variable voltage was connected between the outer electrode and ground of an open-type cell and a cell of later design, an interleaved flat-plate type suitable for high-pressure operation. The potential difference between the electrodes was thus variable, and a plot of the developed voltage versus the "characteristic potential" could be made. The results are shown in Figures 8 and 9. The curves are roughly logarithmic, leveling off at the higher polarizing potentials. These curves illustrate that obtaining the highest possible theoretical value of characteristic potential for a given set of electrodes is not particularly advantageous, because of the slow change in developed voltage with increase in characteristic potential. Characteristic potentials of 0.5 to 1.5 volts, the range we have encountered in various usable cells, give a sufficiently high developed voltage for measurements, and there is at this time little reason for attempting to increase the characteristic potentials.

While the data for Figure 8 were being taken, an important observation was made. The developed voltage was much steadier when the cell was operated with 2 volts across it than when 60 volts were applied to it. Thus, although the developed voltage was about one-half the magnitude with 2 volts that it was with 60 volts, the over-all precision of the



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FIGURE 8

MLM- 629



MLM-629