The amount of C^{14} in the calorimetric samples was estimated from measurements of the density of C^{14}-enriched CO_2 samples relative to that of normal CO_2. The relative density measurements were made by means of a small, extremely sensitive, gas density balance. The volumes of the calorimetric samples were determined by standard methods. The accuracy of the sampling procedure was established by measurements of the relative radioactivity of the various samples.

Chemically pure CO_2 was essential for accurately ascertaining the fraction of $C^{14}O_2$ from the measured relative densities. Active CO_2 was generated from active $BaCO_3$ by thermal decomposition and was purified by repeated fractional distillation. Normal CO_2 was generated from $NaHCO_3$ by thermal decomposition and was similarly purified. After each distillation the relative gas density was measured, and this process was repeated until the density remained constant. The CO_2 was handled in an all-glass system employing mercury cut-off type valves.

The thermal sources employed in the calorimetric determinations were designed to preclude storage of any significant fraction of the total absorbed beta-ray energy. They were made up of approximately 4 ml of the active CO_2 sealed in glass ampoules containing 0.1 g of charcoal and helium gas at a pressure of about 76 cm at room temperature. The ampoules were lined internally with aluminum foil. When the sources were at liquid helium temperatures, the CO_2 was completely adsorbed on the charcoal. Thus, the major portion of the C^{14} beta-rays was absorbed in the charcoal; the remainder was absorbed in the aluminum liner. Storage of absorbed beta-ray energy in metals such as aluminum is highly unlikely. However, the possibility of storage of energy in charcoal at low temperatures was uncertain. Consequently, charcoal was tested for storage of energy by measurements of the powers generated by absorption of Au^{198} beta-rays in aluminum and in charcoal. No storage was detected.

The fraction of $C^{14}O_2$ in the active CO_2 was 0.227, and the active CO_2 generated 3.91×10^{-3} watt per mole. The estimated probable error for each of these experimental values is ±1.0 percent. For pure C^{14}, the power generated per mole is 1.72×10^{-2} watts ±1.4 percent. The product of the decay constant and the average beta-energy of C^{14} is then computed to be 1.79×10^{-10} kev disintegration^{-1} sec^{-1}.

Reported values for the half-life of C^{14} have ranged from less than 5000 to greater than 7000 years. Better agreement exists among recent values, which fall in the approximate range 5400 to 5600 years.[2]

No measurements of the average beta-energy as such have been reported, but the beta-spectrum of the decay has been studied by several investigators.[2] The results are not in agreement as to the exact distribution of the beta-ray energies. Some of these results indicate that the distribution has an allowed shape, but others show deviations from this shape. If the shape of the spectrum is allowed, the average beta-energy can be computed since the maximum energy is known (155 kev). Ketelle[3] has carried out this computation by the method of numerical integration and obtained the value 49.0 kev. Average beta-energy values were also estimated from the published measurements in which an allowed shape was not found. In each case, the estimated value was greater than 49.0 kev.

If the correct value for the average energy is assumed to be 49.0 kev as calculated for a beta-distribution with allowed shape, the results of the present study yield a value of 6030 years for the half-life. Conversely, if the true half-life value is about 5500 years, the average energy calculated from the present results is about 45 kev.

† This work was performed for the AEC.
[1] C. V. Cannon and G. H. Jenks, Rev. Sci. Instr. **21**, 236 (1950).
[2] For references up to 1950, see Hornyak, Lauritsen, Morrison, and Fowler, Revs. Modern Phys. **22**, 333 (1950). Later half-life references are G. G. Manov and L. F. Curtiss, J. Research Natl. Bur. Standards **46**, 328 (1951); A. G. Engelkemeir and W. F. Libby, Rev. Sci. Instr. **21**, 550 (1950); a later reference for the beta-spectrum is S. D. Warshaw, Phys. Rev. **80**, 111 (1950).
[3] B. H. Ketelle, private communication.

Formation and Annulment of Space Charges in Glass and Their Influence on Electric Breakdown

K. J. Keller
Research Department of the N. V. KEMA, Arnhem, Netherlands
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THOUGH it is a well-known fact, the dependence of the electric breakdown field strength of solid dielectrics on the rate of application of stress is often neglected. Previous measurements of the author[1,2] have shown that, in breakdown tests on glass, prebreakdown heating of the test sample is one of the main causes for this dependence, although other effects cannot be excluded. Analogous to the space charges in gases leading to streamer building, it is expected that space charges of ions, left behind by prebreakdown electron avalanches, or of displaced ions in ionic conductors will give rise to field distortion and thus decrease the breakdown voltage with increasing time of application of electric stress.

In recent experiments, it was possible to separate the heat effect from the more permanent space charge effect by (1) prestressing the samples for some seconds with 1.3–1.35 Mv/cm (i.e., for the kind of glass used in these experiments, about 90 percent of breakdown field strength for a time of rise of voltage amounting to 30 seconds), (2) allowing the samples during a few minutes to cool down (if heated) and (3) testing them afterwards with impulse voltage (time of rise 10^{-5} sec, negligible prebreakdown heating). Electrodes consisting of an aqueous solution of $CuSO_4$ were used, as these give the least scattering of breakdown results.

In Fig. 1, N_1 gives the number of impulse breakdown tests with a given breakdown field strength for test specimens, prepared as described in (1), without any pretreatment; N_2 gives the distribution of breakdown field strengths for prestressed samples which show 1 to 3 Mv/cm lower results, owing to field distortion by space charges formed during prestressing. Even after a 20-hour storage at room temperature the effect of prestressing could still be observed. However, prestressed samples that were kept in a furnace at 150°C during 3 hours and cooled slowly showed practically the same distribution (N_3) for impulse tests as the original samples, which proved that prestressing had a reversible effect (space charge) and did not cause permanent damage. N_4 was meant to be a control and showed that the annealing had practically no influence on samples that were not first electrically stressed.

The preceding and other time-dependent effects influencing breakdown field strength measurements should be thoroughly investigated before a theory including these effects can be pro-

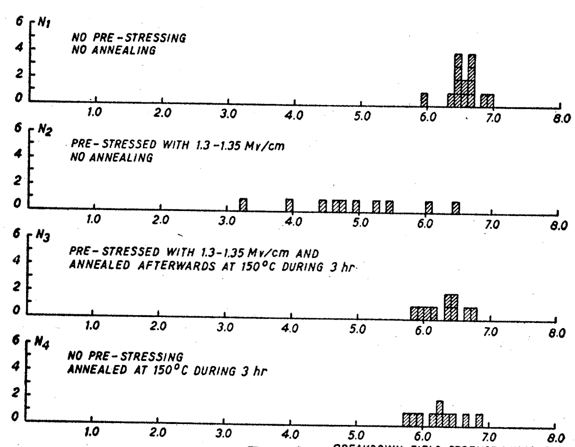

FIG. 1. Histograms showing decrease of breakdown field strength at room temperature of glass caused by space charges generated during prestressing; and removal of the space charges by annealing the test samples after prestressing.

posed, or measurements must be made in which the effects are avoided; to check existing theories with dc experiments is useless.

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[1] K. J. Keller, Physica 14, 475 (1948).
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Recent Studies of the Isotopes of Emanation, Francium, and Radium*

F. F. Momyer, E. K. Hyde, A. Ghiorso, and W. E. Glenn
Ratiation Laboratory and Department of Chemistry, University of California, Berkeley, California
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An earlier study[1] of the alpha-decay characteristics of the low mass isotopes of francium and emanation produced by bombardment of thorium with 340-Mev protons for the purpose of correlating these characteristics with the 126-neutron shell has been continued and expanded to include the element radium.

Greatest progress has been made in the case of emanation where the properties listed in Table I have been measured.

TABLE I. Radioactive properties of low mass emanation isotopes.

Isotope	Observed half-life	Alpha-particle energy ±0.02 Mev	EC/α branching ratio
Em^{209}	31 min	6.02	4–6
Em^{210}[a]	2.7 hr	6.02	~0.1
Em^{211}	16 hr	5.82	2.8
Em^{212}	23 min	6.23[b]	<0.01

[a] This isotope is to be identified with the 2.1-hr activity reported by Ghiorso, Meinke, and Seaborg, Phys. Rev. 76, 1414 (1949).
[b] This value supersedes that given by Hyde *et al.* (see reference 1); also the Fr^{212} alpha-particle energy should be raised to 6.36 Mev.

This work was greatly facilitated by the development of a method for the preparation of platinum plates with the emanation atoms so firmly affixed that counting techniques typical for non-gaseous radioactive samples could be employed. In brief, this method consisted of ionization of the gaseous atoms in a glow discharge and acceleration of these ions into a platinum plate at a potential of a few hundred volts. The method is being applied successfully to krypton and xenon as well as emanation and should be widely applicable in nuclear studies of the nuclides of these elements. This technique resembles that reported by Bergström *et al.*[2] in the study of mass-spectrographically separated radioactive isotopes of rare gas elements.

It is interesting to note that a plot of the alpha-decay energies for the emanation isotopes against neutron number is strikingly similar to the corresponding plot for the isotopes of polonium and astatine as shown in Fig. 1. The alpha-decay energy of At^{211} is given as 5.96 Mev (alpha-particle energy, 5.85 Mev) to correspond with a recent redetermination by us.

Many experiments were performed to obtain information on francium isotopes other than Fr^{212} in this mass region. In this work carrier free fracium fractions were isolated from thorium target solutions by an improved method developed by Hyde.[3] Any isotopes of half-life greater than 5 minutes would have been identified easily. It can be stated that the apparent half-lives of Fr^{213}, Fr^{211}, and isotopes of mass less than 211 are all shorter than 5 minutes. Incomplete results indicate a half-life of 2–5 minutes for Fr^{211} with electron capture (EC) prominent.

Mass assignments in the genetically-linked $Fr^{212}-Em^{212}-At^{208}-Po^{208}$ system were made certain by a mass spectrographic assignment of the key nuclide Fr^{212}. This was done with the time of flight mass spectrometer developed by Glenn.[4]

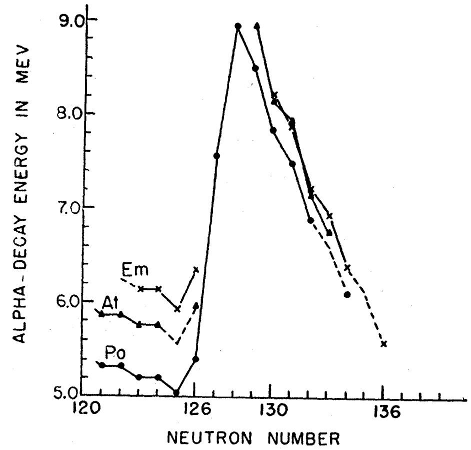

FIG. 1. Similarity of shell effect in elements 84–86 shown by plot of alpha-decay energy *versus* neutron number.

Attempts were made to isolate chemically radium isotopes of mass 214 or less and thus prove that the shell effect extended to this element. However, the stabilization was expected to lengthen the alpha-decay half-lives to the order of only a few minutes. Evidence was found for the following sequence:

$$\underset{\sim 2\text{ min}}{Ra^{213}} \xrightarrow{\alpha} \underset{31\text{ min}}{Em^{209}} \xrightarrow{EC} At^{209}.$$
$$Em^{209} \xrightarrow{\alpha} Po^{205}$$

The Ra^{213} was not observed directly because of its short half-life and because of the interference from heavier radium isotopes.

Cross checks are being carried out using a quite distinct method of preparation of the nuclides, namely the bombardment of lead foils with carbon ions. Miller *et al.*[5] have recently reported the attainment of a sizable beam of energetic (>100 Mev) C^{+6} ions in the Crocker Laboratory 60-inch cyclotron and have effected such reactions as $Au^{197}(C^{12}, 4n)At^{205}$. For our purposes bombardment of lead foils produces radium isotopes of mass 216 or less by such reactions as $Pb^{208}(C^{12}, 4n)Ra^{216}$. These directly produced radium isotopes decay quickly by alpha-particle emission or electron capture to the emanation and francium isotopes in which we are interested. An outstanding advantage of this method, particularly for Ra^{213}, is that none of the higher mass isotopes of these elements can possibly be produced, and hence the interference from them is not present.

As a by-product of the studies of the emanation fraction from the thorium plus proton bombardments, some properties of the previously unreported Em^{221} were observed. The gaseous fraction from the dissolution of a thorium foil target bombarded with 100-Mev protons was purified and placed on a platinum plate using the glow discharge collection technique. This plate when examined in the alpha-ray pulse analyzer showed the alpha-particle peaks corresponding to 4.8-minute Fr^{221} and its 0.020 second daughter At^{217}. Later the expected growth and decay of the Po^{213} alpha-peak were observed. The $Fr^{221}-At^{217}$ double peak decayed with a half-life of 24 minutes. These facts can be interpreted only as meaning that Em^{221} is a beta-emitter of 24 minutes half-life. The alpha-branching is appreciable (of the order of 25 percent) and is currently under investigation.

This work is continuing and a complete report will be issued later.

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