Radioactive Carbon of Long Half-Life

It has been shown by Bonner and Brubaker[1] that C^{14} is unstable relative to N^{14} and is formed by neutrons on nitrogen. Shortly after, McMillan[2] suggested that a soft long-lived radiation found in material removed from the Berkeley cyclotron could be due to C^{14}, formed by deuteron bombardment. Recently Pollard[3] has investigated the nuclear reactions $B^{11}(\alpha,p)C^{14}$ and $C^{13}(D,p)C^{14}$ and found C^{14} to be unstable by no more than 300 kv. He could not detect any activity ascribable to carbon.

The search for long-lived carbon has been under way in this laboratory for some time. Bombardment of an internal ("probe") target[4] of graphite for 120 hr. with 40 μa of 7–8-Mev deuterons has yielded a radioactive body isotopic with carbon. The chemical procedure was similar in many respects to that used by Yost, Ridenour, and Shinohara[5] in the identification of C^{11}.

The graphite target was burned in a stream of O_2 and the gases passed over heated cupric oxide and collected in a liquid-air trap. The gas was absorbed in $Ca(OH)_2$ solution and the resulting $CaCO_3$ filtered and carefully washed. No activity could be detected with a thin (0.1 mm) aluminum solid wall Geiger counter. However a thin film of the $CaCO_3$ ($\sim\frac{1}{3}$ of the total sample) placed inside a screen wall Geiger counter[6] gave $\sim$400 counts/minute. The arrangement was such that by tipping the counter the sample could be made to slide to and fro over the screen in order that the background could be measured alternately with the active sample. When the $CaCO_3$ was converted into CO_2 and introduced into a counter the activity was considerably greater ($\sim$2000 counts/minute). The increase in detectable activity was undoubtedly due in part to more effective solid angle for the soft radiation to be counted and of course self absorption in the sample was completely eliminated.

The gas could be pumped from the counter and the same quantity of ordinary CO_2 introduced with no increase over the background. Indeed, at high partial pressures of CO_2 the counting rate fell below the initial background count since CO_2 is a poor counter gas.

The upper energy limit of the radiation was measured in the screen wall counter by absorption in Al. The maximum energy (assuming negative electrons) appears to be 90,000$\pm$15,000 ev. This is very close to the known value for S^{35}. The above experiments do not exclude S^*O_2. However, sulfur was eliminated by oxidation to SO_4^{--}. The active gas was converted to $CaCO_3$ and then oxidized with $KMnO_4$ in H_2SO_4 solution, $NaHSO_3$ being added as carrier. As an additional precaution the CO_2 was distilled (in vacuum) into $Ca(OH)_2$ again and treated with I_3^- in alkaline solution. After acidification with H_2SO_4 the CO_2 was redistilled into fresh $Ca(OH)_2$. This $CaCO_3$ was radioactive, the recovery being quantitative. Radioactive isotopes of oxygen, nitrogen, and hydrogen are also excluded by the above chemical manipulations.

Stronger samples are in preparation and when available the nature and the energy of the radiation will be more closely examined. A sample has been followed for 10 days and no decay has been detected. The measured cross section coupled with low yield suggests the half-life to be very long (years). Large quantities of nitrogeneous material have been exposed to neutrons for several months and will be shortly worked up for radio-carbon. This latter method should prove more advantageous. It will not only yield higher specific activities of carbon but it will at the same time leave the cyclotron available for other research.

Long-lived radio-carbon will be of great importance for many chemical, biological, and industrial experiments. While the emitted radiation is soft it can nevertheless be used for quantitative work if suitable precautions are taken for the control or elimination of self absorption. The gas technique for counting is by far the most sensitive method.

Experiments are already in progress to extend the results on photosynthesis,[7] etc., obtained thus far with C^{11} (21 min.).

Since the activity is very likely C^{14} formed by $C^{13}{}_6(D^2{}_1,H^1{}_1)C^{14}{}_6$ it is obvious the yields will be increased by bombardment of C^{13} enriched samples. We thank Mr. D. C. DeVault for the use of his circuit in some of these experiments. We are indebted to Professor E. O. Lawrence for his continued encouragement throughout this work. Financial grants to the Radiation Laboratory from the Rockefeller Foundation and the Research Corporation have made these experiments possible.

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February 29, 1940.

[1] L. G. Bonner and G. Brubaker, Phys. Rev. **49**, 778 (1936).
[2] McMillan, Phys. Rev. **49**, 875 (1936).
[3] Pollard, Phys. Rev. **56**, 1168 (1939).
[4] Wilson and Kamen, Phys. Rev. **54**, 1031 (1938).
[5] Yost, Ridenour and Shinohara, J. Chem. Phys. **3**, 133 (1935).
[6] Libby and Lee, Phys. Rev. **55**, 245 (1939).
[7] Ruben, Hassid, Kamen, J. Am. Chem. Soc. **61**, 661 (1939); Ruben, Hassid, Kamen, DeVault, Science **90**, 570 (1939).

A Correction to Measurements with the Electrostatic Analyzer

Allison, Skaggs and Smith[1] have introduced a new method of measuring the energy released in nuclear reactions by deviating the ejected particles through 90° in an electrostatic field. The method is especially suited for particles of low range. A precise measurement of the energy release of the following two reactions was made:

$$Be^9+H^1=Be^8+D^2+Q_1;$$
$$Q_1=0.557\pm0.006 \text{ Mev};$$
$$Be^9+H^1=Li^6+He^4+Q_2;$$
$$Q_2=2.152\pm0.04 \text{ Mev}.$$

On account of the bearing of these results on the range-energy relation and on the evaluation of precise isotopic weights it may be well to point out a correction which has to be applied to them. Allison *et al.* assume that the energy E of the particles liberated in the reaction is not altered by their entrance into the electrostatic analyzer where it is measured (in ev) by $(zV/2)\ln(r_1/r_2)$ where r_1 is the radius of the outer, r_2 the radius of the inner analyzer plate and