the methyl group of methionine contributes insignificantly to the ethanolamine moiety of choline and (b) that the methyl groups of choline and creatine have within experimental limits the same ratio of deuterium to $C^{14}$ as the administered methionine. The present demonstration of the latter point with intramolecular labeling of the methionine methyl groups with deuterium and $C^{14}$ confirms the conclusions drawn in the earlier study[1] with regard to the transfer of methionine methyl as a unit and further shows that hydrogen isotope effects played no significant role in the prior experiment.

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## PRODUCTION OF LABELED ORGANIC MATERIAL WITH ACCELERATED TRITIUM

*Sir:*

Tritium ions when accelerated to an energy of not over 100 e.v. will react to combine with organic material placed in its path. This has led to the production of radioactively labeled organic material and suggests that low energy beams of radioactive atoms may be used as a convenient agent for the practical synthesis of tracer compounds.

These experiments are a consequence of studies of the reactions of recoil tritium atoms produced in nuclear reactions. Recoil tritium will displace hydrogen atoms in organic compounds to substitute in their place and can be used for convenient one-step syntheses of tracer compounds of moderate specific activity. Recent experiments on the mechanism[1,2] of the recoil tritium labeling reaction indicate that the tritons possess only a few electron volts kinetic energy when they undergo reaction to enter organic combination. Thus the 2.7 Mev. possessed by recoil tritons from the $Li^6(n,\alpha)T$ reaction, though useful in enabling the triton to penetrate the material which it is to activate, is expended almost entirely in causing gross radiation damage. To prevent total destruction of the sample by this radiation damage it is necessary to limit the number of recoil tritons and thereby the possible specific activity. Thus by using low energy accelerated tritium rather than high energy recoil tritons it should in principle be possible to produce very high specific activity tracers.

(1) F. S. Rowland, C. N. Turton, and R. Wolfgang, THIS JOURNAL, **78**, 2354 (1956).

(2) R. Wolfgang, J. Eigner and F. S. Rowland, *J. Phys. Chem.*, **60**, 1137 (1956).

A simple discharge tube has been used as the source of accelerated tritium. A cylindrical vessel with electrodes of about 50-mm. diameter about 25 mm. apart was used. About 20 mg. of the material to be activated was spread on the cathode in a thin layer and approximately 0.03–0.06 mm. of $T_2$ introduced. A discharge of about 100 microamp. and about 0.5 hour duration was then passed using a potential difference of the magnitude of 500 volts. The sample which had thus been bombarded with $T^+$ and $T_2^+$ was then removed, subjected to chemical purification to remove gross decomposition products, reduced to gas and counted in a proportional counter. Following this, rigorous further purifications were performed on most of the samples.

Eleven organic compounds ranging in molecular weight from benzoic acid to bovine albumin have been irradiated using this technique. In each case specific activities of the order of 0.1 millicurie/mg. ($\sim 2 \times 10^8$ dis. p. min./mg.) were produced. This tritium is in non-volatile, soluble, non-labile (in water) organic combination. Chromatography indicates that a wide range of labeled species is formed. Some of the activity was incorporated in the compound irradiated but this amount varied erratically from run to run and is less than 10% in most cases and apparently negligibly small in some.

In a control experiment benzoic acid and tritium were placed together but no discharge passed. This yielded some organically bound activity but the amount was smaller by at least a factor of $10^3$ to $10^4$ than if a discharge had been passed.

The incorporation of such a large fraction of the activity in degradation products is probably not anomalous in spite of the expectation that low energy tritium should cause only little total radiation damage. The bombarding tritium has very little penetrating power. This means that all the labeling reactions and all the energy dissipation must proceed within a few monolayers of the surface which may thus become severely damaged. Present work is directed at modifying this effect. Especially if this difficulty is overcome, the use of low energy accelerated radionuclides appears to offer promising possibilities as a simple and broad method for the production of high specific activity tracer compounds of tritium and other radioactive isotopes, *e.g.*, $C^{14}$. It may also provide a fundamental approach to the study of "hot" atoms[3] and ions.

(3) J. Willard, *Ann. Rev. Nuclear Sci.*, **3**, 193 (1953); *Ann. Rev. Phys. Chem.*, **6**, 141 (1955).

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