THE RAPID PREPARATION OF FLUORINE-18 BY MEANS OF NUCLEAR RECOIL SEPARATION

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Abstract

The application of "hot atom chemistry" to the rapid preparation of fluorine-18, t½ = 109.7 min, is described. In this particular case, slurry, or paste targets, of lithium carbonate and distilled water were prepared and subjected to irradiation in the reactor thus:

$$^{6}\mathrm{Li}\,(n,\alpha)\,^{3}\mathrm{H} \qquad ^{16}\mathrm{O}\,(t,n)\,^{18}\mathrm{F}$$

Following the irradiation better than 90% of the fluorine activity produced in the above reaction is found in the water component of the target and 70% of this can be recovered with a single extraction of water. Using a dilute solution of NaOH, containing a trace quantity of fluorine carrier as NaF, yields as high as 98% can be realized

Parameters investigated included yield as a function of Li_2CO_3/H_2O target ratio, and type and temperature of extractant The described method allows the quantitative recovery of the enriched 95% lithium carbonate and, very important, reduces the conventional separation time from 60 - 90 minutes to 7 - 15 minutes depending on the final concentration required for the preparation. Because of its short half-life, fluorine-18, is favoured as an isotope for brain tumor scanning.

Introduction

Fluorine possesses one stable isotope, that of ^{19}F, and four radioisotopes with A = 17 - 21, all of which have short half-lives. The most important of these is ^{18}F, t½ = 109.7 min:β^+, 97%, e-capture, which can be formed by various nuclear reactions, e.g., ^{16}O (α, pn), ^{19}F (n, 2n), ^{20}Ne (d, α), etc. However, for all practical purposes the most important production method can be considered as the ^{16}O (t, n) ^{18}F reaction(1). Using this reaction it is possible to prepare intense carrier free fluorine activity by irradiating lithium carbonate in a reactor, whereby the action of the "hot" triton atom on that of oxygen results in the following:

$$^{6}\mathrm{Li}\,(n,\alpha)\,^{3}\mathrm{H}\,(\mathrm{T}) \qquad ^{16}\mathrm{O}\,(t,n)\,^{18}\mathrm{F}$$

At the present time, ^{18}F, is usually processed by means of co-precipitation(2), distillation(3), absorption on alumina(4), zirconium(5) or magnesium(6), solvent extraction(7) and ion exchange(8). Such chemical processing requires at least one half of the nuclides first half-life(9).

The isolation of ^{18}F by means of nuclear recoil in suspensions of Teflon(10) has been proposed by Amiel and Peisach(11). A similar technique has been reported by Parker for the preparation of carrier free silver using silver colloid suspensions(12). Slurry targets of MgO and MgOH were successfully employed by Parker et al in the production of ^{24}Na through the ^{24}Mg (n, p) ^{24}Na reaction(13).

The work reported in the present communication is based on the slurry target technique, i.e., the suspension of a solid in a suitable liquid, was undertaken in an attempt to reduce as far as possible chemical processing time. This objective has been so well achieved and illustrates so well the purely practical application of hot atom reactions to routine isotope production, that the authors feel that the technique will find extensive application when ever secondary reactions of this kind are involved.

Experimental

For the purpose of the present experiments, 95% enriched lithium-6, in the form of lithium carbonate, was obtained from the Oak Ridge National Laboratory, and carefully weighed out as one gram samples in polyethylene tubes. Barely sufficient distilled water, to completely saturate each Li_2CO_3 sample, was added so as to form a slurry, or paste, in each tube and the latter heat sealed.

The irradiations were carried out in a rabbit facility in a thermal neutron flux of 5×10^{13} $n/cm^2/s$, for a sixty minute period. On completion of the irradiation, a given sample tube was opened and the contents flushed out into a 10 ml beaker with 5 ml of distilled water. In order to realize as complete a transfer as possible of the target slurry, the flushing was achieved at high pressure simply by making use of a 10 ml polyethylene squeez bottle. The target slurry was now subject to vigourous stirring by means of a magnetic stirrer for 5 minutes and then flushed with a further 5 ml of distilled water into the receiving cup of an ultrafiltration assembly fitted with a filter membrane having a pore size of 0.22 μm. Vacuum was applied to the filtration flask and the filtrate collected in a 15 ml centrifuge tube. The filtrate and membrane were carefully removed from the filter assembly for yield determination.

Comparison of the filtrate and membrane activity, i.e., insoluble target material, was made using a Ge/Li detector and multi-channel analyzer and measuring the annihilation gamma-ray of ^{18}F. Calibration was made by means of the 0.662 MeV line of caesium-137. The only activity observed was that of ^{18}F. The samples were also examined for their tritium content by liquid scintillation spectrometry. This proved to be less than 1 μC/ml. Decayed samples (filtrates) were subjected to neutron activation analysis so as to determine the amount of Li_2CO_3 contamination. No such contamination could be observed. Finally, the radiochemical purity of the separated activity was controlled by paper chromatography in the mixture: butanol - methanol - water in the ratio 1 : 3 : 1. The Rf-value for the fluoride ion is 0.53.

Results and Discussion

Irradiations were also carried out with Li_2CO_3 in dry powder form as well as with that prepared as a paste. The target water content was varied within a wide range for the latter form. As could be expected, it was found that with increasing water content of the target for a given irradiation the ^{18}F recoil activity also increased. This effect is shown in fig. 1. It can be seen that with saturation the yield approaches 100%.

Each batch irradiated samples was divided into three making nine samples in all. Recoil activity was extracted from the samples once, twice and three times using cold water, hot (boiling) water and with 0.1 N NaOH containing a trace quantity of NaF (50 μg/ml). The results for the three kinds of extractions are given in tables 1, 2 and 3 respectively. As can be seen from table. 1, 70% of the total fluorine activity produced in the irradiation can be separated from the slurry with a single extraction. The two subsequent extractions contribute at most only an additional 10%. Boiling the water (table 2), during the extraction actually results in a lower initial yield which is no doubt due to volatilization of the free fluoride ion.

As can be seen in table. 3, dilute sodium hydroxide with sodium fluoride carrier results in by far the best extraction efficiency giving a yield of from 88 - 90%. From a routine production point of view, the above figure may be considered so good that it is hardly worth bothering with additional extractions.

The described method allows the rapid preparation of ^{18}F having a radiochemical yield of from 70% - 100% depending on the number of extractions and the use, or not, of carrier.

Obviously, the most important role played in this separation technique is that played by the slurry or paste target, the transition of the "hot atom" from the finely divided solid target to the aqueous phase being irreversible.

In conclusion it is only necessary to add that using recoil separation, preparation time has been reduced from the usual 60 - 90 minutes to a maximum of 15. The product can be obtained without carrier in water solution or with in dilute NaOH. The target material, enriched in ^{6}Li, is uneffected by the entire process and can consequently be used indefinately. The possibility of adapting the technique for use with an in-pile irradiation loop is presently being studied.

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TABLE. 1

Dependence of ^{18}F Yield on Number of Cold Water Extractions

Extraction:	1	2	3
Yield:	70%	78%	79 - 80%

TABLE. 2

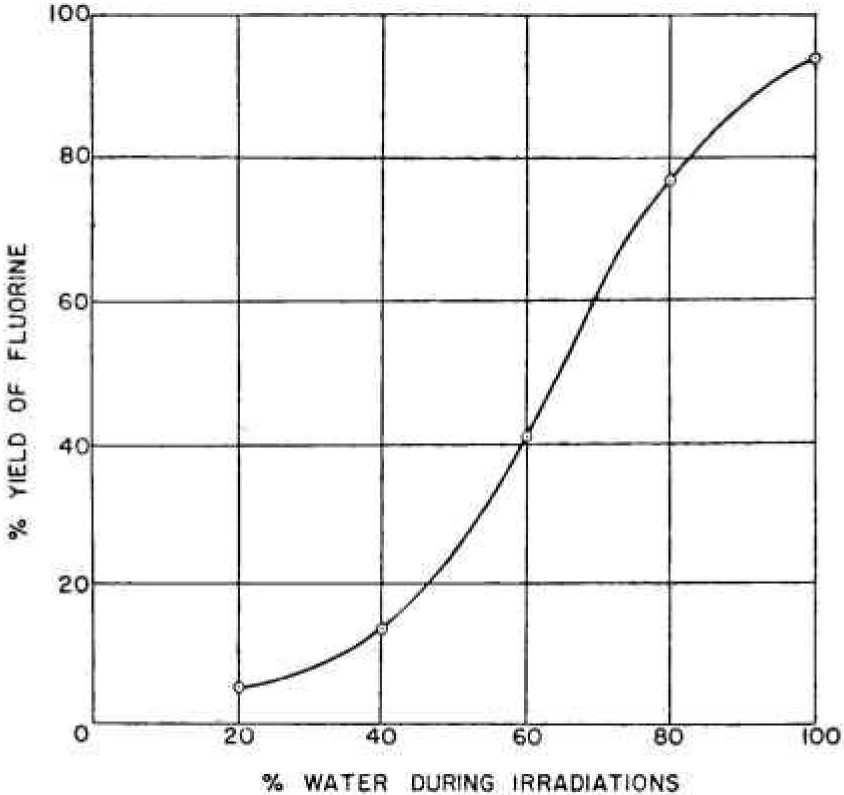
Dependence of ^{18}F Yield on Number of Boiling Water Extractions

Extraction:	1	2	3
Yield:	32%	33 - 34%	34%

TABLE. 3

Dependence of ^{18}F Yield on Number of 0.1 N NaOH + NaF Extractions

Extraction:	1	2	3
Yield:	88 - 91%	95%	98 - 100%


100
80
60
40
20
0
20
40
60
80
100
% YIELD OF FLUORINE
% WATER DURING IRRADIATIONS

Resumo

Descreve-se neste trabalho a aplicação da "química do átomo quente" na preparação do fluôr-18, t½ = 109,7 minutos. Amostras de carbonato de lítio embebidos em água destilada, foram irradiadas no reator IEAR-1, assim:

$$^{6}\text{Li}\ (n, \alpha)^{3}\text{H} \qquad\qquad ^{16}\text{O}\ (t, n)^{18}\text{F}$$

Mais do que 90% da atividade do fluôr produzido na reação acima encontra-se na água do alvo e 70% pode ser recuperado por uma simples extração com água. Usando NaOH diluído contendo traços de carregador de fluoreto de sódio, obtém-se rendimento de 98%.

Os parâmetros estudados incluem o rendimento como função da relação Li_2CO_3/H_2O, o tipo e a temperatura do extractante.

O método descrito permite separação quantitativa do Li_2CO_3 enriquecido a 95% e ainda reduz o tempo de separação convencional de 60 - 90 minutos para 7 - 15 minutos dependendo da concentração final requerida para a preparação.

Devido a meia vida curta do fluôr-18 ele é considerado isótopo útil para cintigrama de tumores cerebrais.

Résumé

On décrit l'application de la "chimie de l'atome chaud" pour la préparation du fluor-18, t½ = 109,7 minutes. Des échantillons de carbonate de lithium en pâte à l'eau ont été irradiées au réacteur IEAR-1, ainsi:

$$^{6}\text{Li}\ (n, \alpha)^{3}\text{H} \qquad\qquad ^{16}\text{O}\ (t, n)^{18}\text{F}$$

Plus que 90% de l'activité du fluor-18 produit, se trouve dans l'eau de la cible irradiée et 70% peut-être récuperée par une extraction à l'eau. Avec le NaOH dilué contenant traces d'entraineur NaF on arrive à un rendement de 98%.

Les paramètres tels que: le rapport Li_2CO_3/H_2O sur le rendement, le type et la température de l'extractant on été etudiés.

La méthode décrite permet une récupération quantitative du Li_2CO_3 enrichi à 95% et réduit le temps de séparation usuel de 60 - 90 minutes à 7 - 15 minutes selon la concentration finale exigée pour la préparation.

Le fluor-18, ayant une demi-periode courte est très utile pour la scintigraphie des tumeurs cérébrales.

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