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The present invention relates to a method of making
enriched radiosotopes and more particularly is directed to
a novel method of producing high specific activity isotopes
in considerably less time than any known process of the
prior art. Such method includes the steps of cation fixa-
tion as an important element thereof.

Until the advent of the present invention the most con-
venient means of obtaining a number of artificial radio-
active elements has consisted of either the separation of
the desired element from fission products or the activation
of a stable nuclei of a suitable element by neutrons
from nuclear reactors. The former process of necessity is
limited to those isotopes occurring in sufficient yields in
the fission product spectrum. The latter process has the
disadvantage that the product nuclei are often isotopic
with the parent material; as a result the specific activities
of such isotopes are necessarily limited and oftentimes can
only be enhanced by prolonged irradiation in a high flux
nuclear reactor.

My invention is in part based upon what may be termed
"cation fixation" and makes use in the preferred embodi-
ment hereof, of the imperfect lattices of substituted alu-
minum silicates. Generally speaking, the present process
involves the introduction of the ionic species desired into
such a lattice structure, the subsequent "fixation" of such
ions, removal of the not-fixed ions, neutron irradiation of
the composite structure and finally the preferential ex-
traction of those nuclei which have gained recoil energy
through the neutron, gamma (hereinafter referred to as
the n,γ) process, and thereby become removed from the
"fixed" sites of the aluminum silicate lattice structures.
The specific details of my invention to enable one skilled
in the art to practice the same are set out below.

One of the more important aspects of my invention is
directed to the production of cobalt[60]. In the conven-
tional process of forming such material, cobalt[59] is placed
in a nuclear reactor where the atoms gain an extra
neutron. As is well known to those skilled in this field
the cobalt must remain in the reactor for several years
in order that cobalt[60] of high specific activity is obtained.
In distinction to such prior art processes by the practice
of the present invention, one is able to make such cobalt[60]
in only several months rather than several years. I
have thus been able to make and segregate useful quanti-
ties of cobalt[60] approximately twenty times faster than
is possible with the conventional processes. Similar en-
hanced reaction and separation times are likewise made
available for a number of other cation species includ-
ing for example, barium, sodium, magnesium, aluminum,
potassium, calcium and scandium.

Furthermore, by the practice of my invention it is
possible to more quickly, conveniently and expeditiously
produce nuclear reaction products which differ in atomic
number from the atomic number of the orginal nuclei
which is placed upon the cation fixation member. Gen-
erally speaking, by such process I can carry out not only
(nuetron, gamma) reactions but other reactions such
as (neutron, proton) and (neutron, alphaparticle) like-
wise. These aspects too of my invention will be con-
sidered in the detail required for the practice of my
process.

Accordingly, a primary object of my invention is to pro-
vide a novel method of making cation radioisotopes which

is considerably faster and more convenient than any
known process of accomplishing a similar result.

Another important object of my invention is to provide
a nuclear reaction process which includes as an impor-
tant step thereof the fixing of nuclei of material to under-
go such reaction on inorganic ion exchange materials,
particularly clays, which are resistant to neutron flux.

A more specific object of my invention is to provide a
new method of making radioactive cobalt, i.e., cobalt[60].

Another more specific object of my invention is to
provide a new method of making radioactive barium,
i.e., barium[139].

Other objects of my invention include the method of
making antimony[125] from tin[124], cobalt[58] from nickel[58]
and scandium[48] from vanadium[51] by various nuclear re-
actions, all of such reactions involving cation fixation.

These and other objects, features and advantages of my
invention will become apparent to those skilled in this
particular art from the following detailed disclosure
thereof.

Before considering some of the specific examples of
the present process, some brief introduction should first
be had to the theoretical principles which underlie it.
As is known, clay minerals, which are a family of natural-
ly occurring aluminum silicates, contain both within
and without their Al—O—Si lattice structure sites for
additional cations. This is what is referred to at times
as imperfect lattice structures and such has been made
use of for years in employing these materials for their
ion exchange capabilities. Under certain circumstances
the cations which are applied to the ion exchange mate-
rial readily exchange and are substituted with other
cations; under other circumstances, I have found that
such cations can be made non-exchangeable or at best,
exchangeable with some considerable difficulty. It is un-
der the latter circumstances that I have been able to fix
the cation in the aluminum silicate clay which is of par-
ticular importance in the practice of my invention.

In the present specification and for reasons of ready
availability in carrying out the experimental processes
which led to my invention the cation fixation means used
was aluminum silicate clays. It will be appreciated that
other similar materials may be employed for this pur-
pose without departing from the spirit or scope of my in-
vention. Such clays were selected both because the
cations being readied for neuclear reaction could be fixed
thereon and because such materials would not undergo
degradation or transformation when subjected to high
density neutron flux. Organic ion exchange materials
should not be used since they do not meet the second
criterion, but certainly other inorganic exchangers may
be substituted in lieu of those specifically named in
the present specification.

For explanatory purposes I wish to next consider a
schematic reaction sequence using for purposes of ex-
ample, naturally occurring sodium clay and any cation,
M^+. The exchangeable and fixed sodium ions are
denoted by $Na^+(. . .)$ and $(Na^+. . .)$ respectively.
Four reactions are of interest to the present process:

(I) $Na(Na—Clay) + M^+ \rightarrow M(Na—Clay) + Na^+$
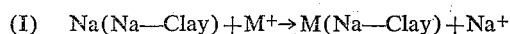
This is the basic exchange reaction by which exchange-
able sodium ions are replaced by M ions. Such reaction
is known to follow the law of mass action and thus

$$[M(Na\text{-}clay)] = \frac{[MNa(Na\text{-}clay][M^+]}{[Na^+]}$$

Accordingly, the replacement can be made substantially
quantitative either by the use of excessive M^+ or the
successive removal of the Na^+ in repeated treatments.

The second reaction of interest involves the fixation of the M^+:

(II)
$$M(\text{Na-clay}) \xrightarrow{\text{Aging}} NaM(\text{M-clay})$$
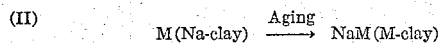
This reaction likewise is an equilibrium process. If necessary the final sample can be made extremely high in fixed M^+ simply by repeated aging and replacement of exchangeable Na^+ with additional M^+. In the case of montmorillonite clays I have found that more than 90% of the originally exchangeable cations can be fixed by heating the clay samples containing the desired cation at 700° C. for approximately 2 to 3 days.

The third reaction is the replacement of the exchangeable Na^+ and M^+ by NH_4^+ or H^+:

(III)
$$NaM(\text{M-clay}) \xrightarrow{NH_4^+} NH_4^+(\text{M-clay})$$
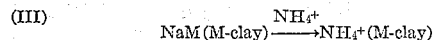
At this stage the M ions are either non-exchangeable or only exchangeable with great difficulty. Such cations are thus quite effectively held within the Al—O—Si lattice structure. Next one should consider the situation when the M^+ nuclei are bombarded with neutrons and undergo an (n,γ) process. The M nuclei would then receive, because of momentum conservation, energy which may be as much as several hundred electron volts and in any event considerably in excess of what is needed to overcome the lattice forces in the Al—O—Si structure. Thus we have the fourth reaction:

(IV)
$$NH_4(\text{M-clay}) \xrightarrow{(n,\gamma)} M^*+NH_4(\ldots \text{clay})$$
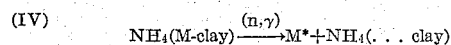
In other words, those M nuclei which have been neutron activated may be projected out of the lattice structure and can therefore be readily separated from the bulk of the M ions which largely remain fixed. It is by the use of the foregoing theoretical consideration in conjunction with the hereinafter laboratory experimentation which gave rise to the present invention.

One aspect of my invention therefore is directed to a modified Szilard-Chalmers process for the preparation of radioisotopes.

My invention may be best understood by consideration of the following examples thereof:

EXAMPLE I

Preparation of cobalt[60]

In this series of experiments the aluminum silicate used was bentonite clay. Sodium-potassium-bentonite, U.S.P. grade was further purified and hydrated by repeated washing with water and recovering the clay by centrifugation. Initially such purified clay was then treated with 300 ml. of N HCl, collected on a Büchner funnel and then repetedly leached with additional amounts of 2 N HCl until no potassium and sodium ions were detectable by the flame test. The combined HCl solutions containing the exchangeable potassium and sodium ions were analyzed by flame photometry to determine the cation exchange capacity of such clay to be 79.7 millequivalence per 100 grams of clay.

Another aliquot of the purified and hydrated sodium-potassium-bentonite was contacted with dilute aqueous solution of cobaltous chloride, collected on a Büchner funnel and repeatedly leached with $CoCl_2$ solution until the filtrate was free of sodium and potassium ions as detected by the flame test. This Co-clay was then dried and calcined at approximately 700–720° C. for periods ranging from 48 to 72 hours to effect the cation-fixation. Subsequently the clay was treated to remove as much as possible of the exchangeable cobalt contained therein. This step of the process was accomplished either by leaching with aqueous HCl solution or electrolytic extraction. In leaching a predetermined volume of 0.02 N HCl containing H^+ in a millequivalent amount twice as large as that of the Co^{++} present in the specific bentonite sample was employed.

The samples were then irradiated either in the Armour

Research Foundation boiling water reactor or in the CP–5 reactor of the Argonne National Laboratory. In all cases referenced cobalt standards were irradiated with the clay samples to provide information concerning any possible fluxations in the integrated neutron fluxes.

EXAMPLE II

Preparation of barium[139] from barium[138]

In this process the treatment steps are very similar to those which are set out in the example presented above. The same type of clay was used and was purified in the same manner as in the cobalt experiments. In this case the clay samples were treated with a dilute solution of $BaCl_2$, collected on a Büchner funnel and repeatedly leached with additional amounts of $BaCl_2$ solution until the filtrate contained no sodium or potassium ions detectable by flame tests. The amount of barium ions introduced based on that of sodium and potassium ions exchanged and as determined by flame photometry, was found to be 68.3 millequivalents per 100 grams of clay. Finally the barium bentonite obtained was heated for 48 hours at 700° C. in order to accelerate the barium ion fixation onto the clay. The extent of the Ba^{++} ion fixation in these clay samples, based upon the amount of Ba ions, subsequently eluted with HCl amounted to 76.9% of the Ba ions originally introduced into the sample.

The fixed barium-bentonite clays were then irradiated for time periods ranging from 20 to 25 minutes in the ARF reactor where the neutron flux is 6×10^{11} neutrons cm.$^{-2}$ sec.$^{-1}$. In this case $BaCO_3$ was used as the reference standard.

Following irradiation the barium-bentonite samples were eluted twice with dilute solutions of either HCl or CH_3COONH_4 of predetermined concentrations. Following such treatments the supernatant liquids containing the released Ba ion were separated by centrifugation and decantation, concentrated by evaporization and then transferred into a glass counting vessel (containing approximately 0.2 gram of NH_4NO_3 which was added to aid subsequent handling) and finally evaporated to dryness. The dried samples containing the barium[139] isotope were measured by means of an RCl 256 channel differential pulse height analyzer using a 3 x 3 inch NaI (Tl-activated) crystal.

Enrichment factors were determined for both the cobalt and barium samples.

As noted above, following irradiation the nuclei produced were extracted. This was accomplished either by ion exchange procedures (i.e., treating the irradiated clay samples with dilute solutions of HCl or NH_4Cl) or by electrodialysis (i.e., electrolyzing aqueous suspensions of the irradiated samples and collecting the radioactive nuclei in the cathodic chamber).

Following this determination of the enrichment factor was carried out by neutron-activation analysis for the presence of stable cobalt-59 or barium-138 nuclei in the extracted sample using the formula

$$\text{Enrichment factor} = \frac{\text{Spec. activity of the extract}}{\text{Spec. activity if no enrichment}}$$

$$= \frac{C_1}{C_2 - C_1} \frac{nvt \text{ (re-exp.)}}{nvt \text{ (original)}}$$

for the Co-60 samples, or
$$\frac{C_1}{C_2} \frac{nvt \text{ (re-exp.)}}{nvt \text{ (original)}}$$

for the Ba-138 samples. (In these formulae, C_1 and C_2 represent respectively the counting rate of a sample before and after re-exposure for neutron activation analysis, whereas nvt (re-exp.) and nvt (original) represent respectively the integrated fluxes used in the activation analysis and in the original hot-atom experiment. The difference between the formula for Co-60 and that for Ba-139 is due to the short half life of the latter nuclei).

The results of our experiments showed that the enrichment factors achieved are approximately 20 or higher

in the case of Co-60, and approximately 3 in the case of Ba-139. The enrichment factors achieved are not sensitively affected by the intensity of the neutron flux over some three orders of magnitude, but are dependent to a

considerable extent on the prior treatment of the clay samples as well as on the method of extraction.

The following tables present some of the results of the hereinabove experiments.

TABLE 1.—COUNTING DATA AND ENRICHMENT FACTORS SELECTED COBALT-60 CHEMICAL ELUTION EXPERIMENTS

Sample Identification	Counting Data (c.p.m.), Original (C_0)	Re-exposure (C_1)	$\frac{(nvt)\ (re\text{-}exp.)}{(nvt)\ (original)}$	Enrichment Factor
M–Feb 3–46–47	1.337×10^4	1.445×10^4	3.92	49
M–Fin 3–58–59–a	4.052×10^3	4.348×10^3	3.43	47
M–Fin 3–58–59–b	1.618×10^3	1.804×10^3	3.43	30
M–Fin 3–60–61–a	0.830×10^3	0.946×10^3	2.94	21
M–Fin 3–60–a	1.312×10^3	1.488×10^3	2.39	18
M–Jan 3–2	9.39×10^3	9.92×10^3	1.0	18
M–Jan 3–1	2.20×10^4	2.36×10^4	1.0	14
M–Fin 3–56–57–a	7.148×10^3	8.982×10^3	3.04	12
M–Fin 3–56–57–b	3.154×10^3	4.129×10^3	3.04	10
M–Dec 2–1	0.964×10^3	1.096×10^3	0.97	7.3
M–Feb 3–2	1.08×10^4	2.05×10^4	3.92	4.4
M–Dec 2–2	0.469×10^3	0.583×10^3	0.99	3.0

TABLE 2.—COUNTING DATA AND ENRICHMENT FACTORS SELECTED COBALT-60 ELECTROCHEMICAL ELUTION EXPERIMENTS

Sample Identification	Counting Data (c.p.m.), Original (C_0)	Re-exposure (C_1)	$\frac{(nvt)\ (re\text{-}exp.)}{(nvt)\ (original)}$	Enrichment Factor
V–Feb 3–11–1	1.10×10^3	1.46×10^3	6.5	20
V–Feb 3–11–2	1.11×10^3	1.57×10^3	6.5	16
V–Nov 2–7	7.46×10^3	11.7×10^3	1.0	1.7

TABLE 3.—EFFECT OF ELUENT CONCENTRATION

Sample Identification	Effluent Number	Counting Data (c.p.m.) Original	Counting Data (c.p.m.) Re-exposure [1]	Percent Yield [2]	HCl Concentration	Enrichment Factor
V–5–1	1	5.10×10^3	8.89×10^3	2.98	0.00296 N	6.50
	2	3.13×10^3	5.53×10^3	1.82		6.27
	3	1.51×10^3	2.71×10^3	0.88		6.03
V–5–2	1	8.85×10^3	15.24×10^3	3.52	0.00592 N	6.65
	2	4.40×10^3	7.85×10^3	1.76		6.16
	3	2.89×10^3	5.24×10^3	0.89		5.91
V–5–3	1	7.71×10^3	13.35×10^3	3.76	0.0118 N	6.58
	2	2.89×10^3	5.24×10^3	1.41		5.95
	3	1.67×10^3	3.10×10^3	0.82		5.65
V–5–4	1	6.58×10^3	11.05×10^3	3.43	0.0178 N	7.08
	2	3.85×10^3	6.89×10^3	2.00		6.08
	3	1.51×10^3	2.83×10^3	0.79		5.46
V–5–5	1	10.55×1^3	18.17×10^3	4.07	0.0237 N	6.66
	2	5.79×10^3	10.29×10^3	2.24		6.17
	3	1.71×10^3	3.11×10^3	0.66		5.90
V–5–6	1	15.44×10^3	26.49×10^3	5.87	0.0296 N	6.74
	2	6.18×10^3	11.10×10^3	2.35		6.02
	3	2.11×10^3	3.95×10^3	0.81		5.52

[1] nvt (re-exp.)/nvt (orignial) = 4.81.
[2] Percent of radionuclide formed.

TABLE 4.—EFFECT OF TIME ELAPSED BETWEEN IRRADIATION AND PROCESSING

Sample Identification	Days Elapsed	Effluent Number	Counting Data (c.p.m.) Original	Counting Data (c.p.m.) Re-exposure [1]	Percent Yield [2]	Enrichment Factor
V–6–1	1	1	4.82×10^3	8.56×10^3	3.0	6.4
		2	1.98×10^3	3.62×10^3		6.1
		3	1.14×10^3	2.01×10^3		6.6
V–6–2	2	1	5.97×10^3	10.47×10^3	3.0	6.6
		2	2.07×10^3	3.79×10^3		6.0
		3	1.15×10^3	2.04×10^3		6.4
V–6–3	5	1	5.54×10^3	9.87×10^3	2.7	6.4
		2	1.92×10^3	3.54×10^3		6.0
		3	1.50×10^3	2.76×10^3		5.9
V–6–4	9	1	1.15×10^3	2.09×10^3	1.1	4.6
		2	0.95×10^3	1.00×10^3		5.6
		3	3.18×10^3	5.40×10^3		7.1
V–6–5	16	1	6.21×10^3	10.93×10^3	2.9	6.6
		2	2.40×10^3	4.36×10^3		6.2
		3	1.42×10^3	2.51×10^3		6.6

[1] nvt (re-exp)/nvt(original) = 5.0.
[2] Percent of radionuclide formed.

TABLE 5.—EFFECT OF INTEGRATED NEUTRON FLUX [1]

Sample Identification	Exposure Time (days)	Effluent Number	Counting Data (c.p.m.)		$\frac{nvt \text{ (re-exp.)}}{nvt \text{ (original)}}$	Enrichment Factor
			Original	Re-exposure		
V-4-1	1	1	422	1,068	10	6.6
		2	434	1,290	10	5.1
V-4-2	2	1	1,148	1,795	5	7.9
		2	1,017	1,606	5	8.6
V-4-3	3	1	1,693	2,450	3.33	7.5
		2	645	940	3.33	7.4
V-4-4	4	1	2,077	2,645	2.5	10.1
		2	1,270	1,660	2.5	8.5

[1] It is to be noted that enrichment factors actually appear to become higher with increasing integrated flux. This could be due to the diminishing relative importance of the residual exchangeable cobalt-59 ions remaining in the system as the amount of Co-60 increase.

TABLE 6.—COUNTING DATA AND ENRICHMENT FACTORS—BARIUM EXPERIMENTS

Sample Identification Numbers [1]	Zero Time Counting Rates (c.p.m.)				Ba Standards (mg.)		Enrichment factor $\frac{ADE}{CBF}$
	Eluted Ba-138 and Ba-139 (A)	Ba Standard Irradiated with clay (B)	Eluted Ba-139 after re-irradiation (C)	Ba Standard used in re-irradiation (D)	With original sample (E)	With eluted Ba-139 (F)	
M16-17	3.05×10^4	6.50×10^5	1.07×10^4	5.80×10^5	6.20	5.08	3.10
M24-25	6.39×10^3	4.25×10^5	3.08×10^3	1.95×10^5	1.95	0.975	1.90
M26-27	1.91×10^4	3.05×10^5	8.00×10^3	3.10×10^5	0.835	0.766	2.64
M28-29	2.72×10^4	7.70×10^5	9.74×10^3	1.08×10^6	0.766	1.04	2.88
M30-31	1.68×10^4	1.76×10^6	6.47×10^3	7.40×10^5	1.46	0.766	2.08

[1] Two numbers are used for each set of experiments. One is for the original sample, the other for the re-irradiated elution product.

It will be understood that various modifications and variations may be effected without departing from the spirit or scope of the novel concepts of my invention.

I claim as my invention:

1. The method of making radioisotopes which comprises the steps of: introducing cations to be made radioactive to an inorganic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such cations within its lattice structure; fixing said cations in said cation exchange material; removing not-fixed cations; irradiating said ion exchange material-fixed cation composite with neutrons to produce radioactive product nuclei of said cations and concurrently defixing such radioactive product nuclei; and separating such radioactive nuclei from the ion exchange mass.

2. The method of carrying out a nuclear reaction which comprises the steps of: introducing cations to be made radioactive to an inorganic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such cations within its lattice structure; fixing said cations in said cation exchange material; removing not-fixed cations; irradiating said ion exchange material-fixed cation composite with nuclear radiation to produce radioactive product nuclei of said cations and concurrently defixing such radioactive product nuclei, and separating such radioactive nuclei from the ion exchange mass.

3. The method of making radioisotopes which comprises the steps of: introducing cations to be made radioactive to an aluminum silicate clay ion exchanger material which is characterized by a closed lattice structure; fixing said cations in said exchanger; removing not-fixed cations; irradiating said exchanger-fixed cation composite with neutrons to produce a radioactive isotope of said cations and concurrently defining such cations and separating said cations from the exchanger mass.

4. The method of making cobalt60 which comprises the steps of: introducing ions of cobalt59 to an inorganic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such ions within its lattice structure; heating said cobalt59 containing ion exchange material at approximately 700° C. for at least 24 hours to fix said ions in said cation exchange material; removing not-fixed barium; irradiating said ion exchange material-cobalt59 composite with neutrons to produce cobalt60 and concurrently defixing such cobalt60, and separating such cobalt60 from the ion exchange mass.

5. The method as defined by claim 4 wherein said inorganic cation exchange material is an aluminum silicate clay.

6. The method of making barium139 which comprises the steps of: introducing ions of barium138 to an inorganic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such ions within its lattice structure; heating barium138 containing ion exchange material at approximately 700° C. for at least 24 hours to fix said ions in said cation exchange material; removing not-fixed cobalt ions; irradiating said ion exchange material-cobalt59 composite with neutrons to produce cobalt60 and concurrently defixing such cobalt60, and separating such cobalt60 from the ion exchange mass.

7. The method as defined by claim 6 wherein said inorganic cation exchange material is an aluminum silicate clay.

8. The method of making cobalt60 which comprises the steps of: introducing ions of cobalt59 to an inorganic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such ions within its lattice structure; fixing said cobalt59 ions in said cation exchange material; removing not-fixed cobalt; irradiating said ion exchange material-cobalt59 composite with neutrons to produce cobalt60 and concurrently defix such cobalt60, and separating such cobalt60 from the ion exchange mass.

9. The method of making barium139 which comprises the steps of: introducing ions of barium138 to an in-

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organic cation exchange material which is resistant to neutrons and other ionizing radiation and which is further characterized by the ability to fix such ions within its lattice structure; fixing said barium[138] on said cation exchange material; removing not-fixed barium; irradiating said ion exchange material-barium[138] composite with neutrons to produce barium[139] and concurrently defix such barium[139], and separating such barium[139] from the ion exchange mass.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,167,479   January 26, 1965

Paul Y. Feng

It is hereby certified that error appears in the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 8, line 41, for "barium" read -- cobalt --; line 56, for "cobalt" read -- barium --; line 57, for "cobalt^{59}" read -- barium^{138} --; same line 57 and lines 58 and 59, for "cobalt^{60}", each occurrence, read -- barium^{139} --.

Signed and sealed this 25th day of February 1969.

(SEAL)
Attest:

Edward M. Fletcher, Jr.   EDWARD J. BRENNER

Attesting Officer   Commissioner of Patents

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