A ^{82}Sr-^{82}Rb ISOTOPE GENERATOR FOR USE IN NUCLEAR MEDICINE

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An improved ^{82}Sr–^{82}Rb generator system, based on the complexing ion-exchange resin Chelex-100, has been developed. Columns of this material can be easily and rapidly milked, and the rubidium–strontium separation factor for a fresh generator under the experimental conditions studied was found to be $>10^7$. Approximately 80% of the ^{82}Rb present can be delivered in a 15-ml volume of aqueous 0.2 M NH_4Cl solution. After more than 6 liters of eluant had passed through the generator, the rubidium–strontium separation factor was still observed to be $>10^5$ and no unusual strontium breakthrough behavior was seen in the system over nearly three ^{82}Sr half-lives.

Full-scale operation of the Clinton P. Anderson Meson Physics Facility (designated LAMPF) at the Los Alamos Scientific Laboratory will provide significant quantities of 25-day ^{82}Sr for clinical investigation (*1*). The short-lived daughter, 75-sec ^{82}Rb, could prove of value in biomedicine for circulation and perfusion studies (*2*) as well as for myocardial imaging (*3*). A radiochemical separation procedure for the quantitative recovery and purification of spallation-produced ^{82}Sr from proton-irradiated molybdenum targets has recently been developed (*4*), and the identities and decay properties of the activities present in the final chemical process solution are given in Table 1.

The existence of a suitable ^{82}Sr–^{82}Rb isotope generator is crucial to the utility of this radionuclidic system in nuclear medicine. Although many effective strontium–rubidium separations have been implemented in such diverse fields as fission research (*9*), geochemical and cosmochemical chronology studies (*10*), and isotope production (*11*), few methods satisfy the following stringent requirements of a potential biomedical radionuclide generator:

1. The system should be simple to operate.
2. Near-quantitative ^{82}Rb yields should be obtained from the generator with each milking to maximize the system efficiency.
3. The generator must have extremely low strontium breakthrough per elution to minimize the amount of long-lived, bone-seeking radiostrontium activities administered to the patient. Conditions 2 and 3 taken together imply a large rubidium–strontium separation factor.

Received Aug. 29, 1974; revision accepted Nov. 18, 1974.

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TABLE 1. RADIONUCLIDES PRESENT FOLLOWING CHEMICAL SEPARATION SCHEME FOR ^{82}Sr FROM Mo METAL*

Nuclide	Half-life	Decay modes	Major E_γ in keV (intensity in %)	References
^{82}Rb	1.25 min	94.6% β^+ 5.4% EC	511.0 (189%, $\gamma^\pm$) 776.6 (13.4%) 1,395.2 (0.47%)	(5,6,7)
^{82}Sr	25.0 days	100% EC	None	(5,6)
^{85}Sr	64.5 days	100% EC	514.0 (99.3%)	(8)

* See Ref. 4 for complete details.

4. The generator milking time should be short in comparison with the ^{82}Rb half-life. This will keep the amount of in situ ^{82}Rb decay small and therefore the effective overall ^{82}Rb yield high.
5. The generator eluant must be compatible with, or have the potential to be easily and rapidly made compatible with, biologic systems. The very short half-life of ^{82}Rb precludes the performance of any detailed post-elution chemistry in the interest of efficient radiorubidium yields.
6. The system should have sufficient stability on a time scale of several ^{82}Sr half-lives to allow repetitive usage and a reasonable shelf life.

The only ^{82}Sr–^{82}Rb biomedical generators of which we are aware are those developed at Donner Laboratory (*3*) as derived from previous work performed at Oak Ridge (*11*). These systems employ the weakly acidic cation-exchange resin Bio-Rex 70, carrier-free ^{82}Sr, and an automatic elution system for intravenous infusion. One generator uses varying strengths of ammonium acetate ($NH_4C_2H_3O_2$) solution as the eluant, but it is restricted to concentrations ≤ 0.4 *M* because of the toxicity of the acetate compound. The rubidium-strontium separation factor for a fresh generator is 10^4, but passage of 400 ml of 0.3 *M* $NH_4C_2H_3O_2$ through the column reduces this value to 10^2, and the ^{82}Rb yield in a 20-ml elution is only 56%. Another generator elutes the ^{82}Sr-loaded column with a 3% NaCl solution. This system exhibits a 10^5 maximum rubidium–strontium separation factor, no significant increase in strontium leakage with up to 600 ml of eluant, and a ^{82}Rb elution yield of 62%.

We have improved on the Berkeley generators by making use of the chemical fact that the alkali metal elements rarely, if ever, form coordination complexes (*12*). Moreover, previous work on the retention of calcium on the chelating exchanger Dowex A-1 demonstrated that distribution coefficients $>10^4$ could be obtained for alkaline earths in solutions of high pH and low ionic strength (*13*). The behavioral similarity of calcium and strontium on a chelating resin (*14*) as well as the expectation of a lack of rubidium interaction led to the development of a new radioisotope generator based on the ion-exchange resin Chelex-100.

EXPERIMENTAL

Chemistry. Three independent ion-exchange column experiments were performed. In each, a glass column of 1.1 cm i.d. was filled to a height of approximately 6-6.5 cm with 100-200 mesh Chelex-100 analytical-grade resin obtained from Bio-Rad Laboratories. The resin was slurried into the columns with a pH 9.3–9.4 buffer solution of 0.1 *M* NH_4OH + 0.1 *M* NH_4Cl, and this same solution was used as the generator eluant for the subsequent milking of ^{82}Rb. The flow rate for column loadings was always approximately 0.5–1 ml/min.

For the first experiment, the weakly acidic final solutions from several Mo–^{82}Sr radiochemical separations (*4*) were combined, adjusted to pH of about 9.5 with conc NH_4OH, and diluted to 100-150 ml with distilled water. This solution was then charged onto a Chelex-100 column. Successive elutions were performed with the NH_4OH–NH_4Cl buffer at a flow rate of about 1 ml/sec, and, as seen from a typical elution curve (Fig. 1), a 25-ml eluant volume was found to be sufficient for quantitative ^{82}Rb milkings under these experimental conditions. A total of 2,600 ml was passed through this column to determine the strontium breakthrough characteristics, with 20 independent 25-ml eluant volumes being sampled at various points to measure ^{82}Rb yields. The radiostrontium activities present for this investigation were assayed to be approximately 0.5 μCi ^{82}Sr and 5 μCi ^{85}Sr.

To determine more realistically the strontium breakthrough for this generator system, a second experiment was performed in which 10 mCi of commercially obtained ^{85}Sr was introduced onto a fresh Chelex column (again, after pH adjustment to approximately 9.5 and dilution). More than 6 liters of the eluant buffer were passed through the resin at flow rates of 0.6-0.8 ml/sec, and 25-ml volumes were collected periodically to measure their radiostrontium content.

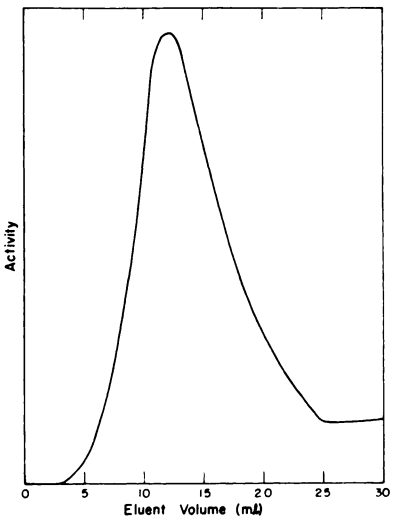

FIG. 1. Rubidium-82 elution from 100-200 mesh Chelex-100 ion-exchange column. Eluant is 0.1 *M* NH_4OH + 0.1 *M* NH_4Cl buffer solution; column dimensions are 1.1 cm i.d., 6-6.5 cm length; flow rate equals approximately 1 ml/sec.

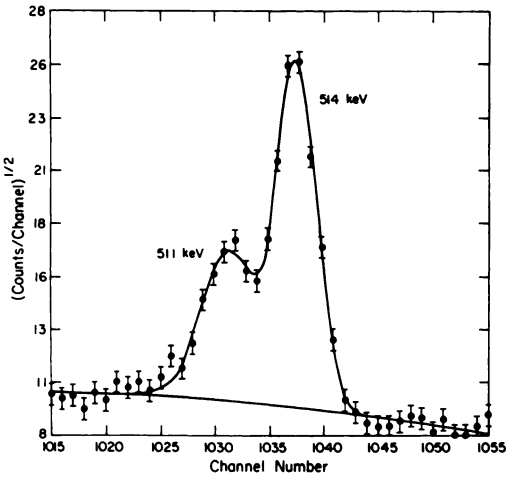

FIG. 2. Ge(Li) spectrometer system and computer resolution of 511 ($\gamma^{\pm}$) and 514-keV gamma rays.

Finally, to study the long-term stability of this system, the ^{85}Sr was stripped from the previous generator with a few column volumes of 1 *M* HCl, the solution was made alkaline and dilute, and the ^{85}Sr was reintroduced onto another freshly made Chelex column. This new generator was then milked intermittently for several weeks to determine any unusual time-dependent behavior of the strontium breakthrough. By the conclusion of this experiment, the quantity of ^{85}Sr present on the column had decayed to approximately 4 mCi.

Radioactivity analysis. The various 25-ml samples of the column elutions were fixed in a constant, reproducible geometry in polyethylene containers and counted on the Ge(Li) spectrometer system described previously (*4*). Gamma spectra were recorded on magnetic tape and subsequently analyzed on a CDC 7600 computer with both of the program codes, SAMPO (*15*) and GAMANAL (*16*). Rubidium-82 behavior was monitored through the 511 ($\gamma^{\pm}$) and 777-keV gamma rays whereas strontium breakthrough was measured by the ^{85}Sr 514-keV transition. The capabilities of the gamma spectrometer and computer codes were such that the 511-keV (Doppler-broadened) and 514-keV photopeaks were readily resolved and Fig. 2 depicts typical counting data in this energy region along with the computer fit to that data.

RESULTS AND DISCUSSION

In the first column experiment the 20 independent elutions to measure ^{82}Rb yield gave an average value of 102 ± 3% radiorubidium off the column in a 25-ml volume. The measured ^{82}Rb counting data were decay-corrected to the start of elution to obtain this percentage, however, and the practical ^{82}Rb generator yield (the amount capable of being administered to a patient) must also reflect the decay of the isotope during transit of the column. From Fig. 1, it was determined that 90–95% of the total activity can be found in the 15-ml eluant volume between 5 and 20 ml. At a flow rate of 1 ml/sec, therefore, it will take 20 sec to pass 20 ml through the generator and this will give rise to a 17% ^{82}Rb decay factor. As a result, the effective ^{82}Rb yield from this column would be approximately 80%.

The strontium breakthrough results for this first experiment varied from 10^{-7} to 10^{-3} while the rubidium–strontium separation factor ranged from 10^{6} to 10^{2} [separation factor = (fraction of strontium breakthrough)$^{-1}$ since the rubidium removal is 100%]. Because this column employed the combined final solutions from several Mo–^{82}Sr radiochemical separations (*4*), there were approximately 20 mg of stable strontium carrier present in the solution charged onto the generator. This large column loading will not be typical of the LAMPF-generated ^{82}Sr product although a milligram or so of stable strontium isotopes will probably be produced by spallation mechanisms under the intense irradiation conditions of future LAMPF production runs. Consequently, the strontium breakthrough data of the first experiment should not be considered typical of the true performance of the Chelex generator under more realistic column-loading parameters (the ^{82}Rb yield results are valid, however).

A better indication of the actual strontium breakthrough characteristics for a more likely situation was obtained in the second column experiment. The 10 mCi of commercially produced ^{85}Sr contained approximately 0.8 mg of carrier strontium, an amount much closer to the eventual LAMPF product than the 20-mg quantity of the first column. The results of this investigation are presented in Table 2. The rubidium–strontium separation factor for a fresh generator was observed to be of the order of 10^{7} and, even after more than 6 liters of eluant had been passed through the column, this variable was still $>10^{5}$.

The results of the long-term radiation damage study are shown in Fig. 3. Over a period of nearly three ^{82}Sr half-lives, no perceptible deviation of the strontium breakthrough from a linear behavior was noted.

CONCLUSIONS

Chelex-100 resin has been used as the basis of a new ^{82}Sr–^{82}Rb isotope generator. Under the experimental conditions described in this paper, the rubidium–strontium separation factor for a fresh system is $>10^{7}$, and the useful ^{82}Rb yield off the column is approximately 80%. A postelution neutralization

TABLE 2. STRONTIUM BREAKTHROUGH AND RUBIDIUM–STRONTIUM SEPARATION FACTORS FOR CHELEX-100 GENERATOR WITH 0.8 mg STRONTIUM COLUMN LOADING

Volume through column (ml)	Sr breakthrough/ 25-ml elution	Rubidium–strontium separation factor
450	2.5×10^{-7}	4.0×10^{6}
700	1.1×10^{-7}	9.3×10^{6}
950	6.8×10^{-8}	1.5×10^{7}
1,200	5.6×10^{-8}	1.8×10^{7}
1,450	4.9×10^{-8}	2.0×10^{7}
1,700	4.0×10^{-8}	2.5×10^{7}
1,950	5.1×10^{-8}	2.0×10^{7}
2,200	3.5×10^{-8}	2.8×10^{7}
2,700	1.4×10^{-7}	7.2×10^{6}
3,200	7.8×10^{-8}	1.3×10^{7}
3,700	9.1×10^{-8}	1.1×10^{7}
4,200	2.2×10^{-7}	4.6×10^{6}
4,700	3.5×10^{-7}	2.9×10^{6}
5,200	7.0×10^{-7}	1.4×10^{6}
5,700	1.6×10^{-6}	6.4×10^{5}
6,200	3.6×10^{-6}	2.7×10^{5}

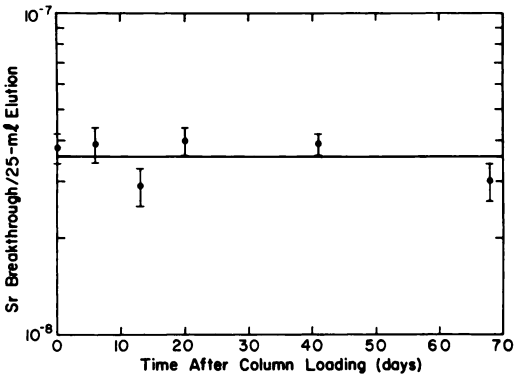

FIG. 3. Strontium breakthrough determined as a function of time after column loading.

of the eluant with a small volume of a concentrated HCl solution would make the ^{82}Rb-containing fluid more physiologically tolerable and would allow injection of essentially a 0.2 *M* NH_4Cl solution. The generator milking is rapid, repetitive, and easy to perform. In accordance with the laws of radioactive secular equilibrium (*17*), quantitative ^{82}Rb elutions can be performed every 10 min or so.

More than 6 liters of eluant could be passed through the system described here without decreasing the rubidium–strontium separation factor below 10^5. Should strontium breakthrough become unacceptable, however, it is a simple procedure to quantitatively strip the radiostrontium from the resin with a few column volumes of 1 *M* HCl, adjust the pH and ionic strength as discussed before, and prepare a fresh Chelex generator. In this regard, one should be aware of the cautions concerning Chelex-100 swelling and the storing of the resin in the hydrogen form (*14,18*).

In comparing our results with the performance of other ^{82}Sr–^{82}Rb generators (*3*), it should be remembered that previous work employed carrier-free ^{82}Sr whereas this study utilized a minimum of 0.8 mg of stable strontium. It is expected that the performance characteristics of our macroscopically loaded column experiments would be considerably improved if conducted in the carrier-free mode.

We hope that the results presented here will serve as a starting point for further research on Chelex-based generators by other investigators. System parameters such as strontium breakthrough and delivery volume are very sensitive to adjustable variables like column dimensions, flow rate, resin size, temperature, and, for chelating resins, pH (*19,20*). For example, employing longer and thinner columns, slower flow rates, eluants with a higher pH, or perhaps a mixed water–ethanol medium (*21*) may improve the strontium breakthrough characteristics. Individual investigators could then easily design systems to meet their own specific requirements of ^{82}Rb yield, delivery volume, etc.

ACKNOWLEDGMENTS

This work was performed under the auspices of the United States Atomic Energy Commission and was supported in part by American Cancer Society Research Grant DT-23A.

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