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Informal

FEB 15 1967

MILKING SYSTEMS - STATUS OF THE ART

MASTER

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Milking systems as means of separating a desired radio-isotope from its parent and other contaminants have proved to be extremely useful. The milking system, or "generator", incorporates a relatively long-lived parent activity, fixed usually in a solid form, from which its shorter-lived daughter can be eluted from the parent phase as required. In this way relatively short-lived isotopes may be stored at the place of use. The system must, however, meet certain requirements in order to be of real value. As used at Brookhaven, it must be simple to handle, operate, and shield. It must also yield rapidly a product of excellent purity in a chemical form suitable for use with little or no treatment. The parent material must remain in a form or be readily reconverted into a form from which the process can be repeated many times.

Many systems have been proposed, some of which have proved practical and some of which have not yet proved useful.

One of the most recently tried media for the successful separation of carrier-free radionuclides is Vycor glass. The

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powder has been tried in column form as an adsorbent for niobium-95. Zirconium-95 was successfully eluted with 10 M HNO_3 and was about 99% pure. (1)

R. Münze has developed a generator for the continuous separation of carrier-free iodine-132 from tellurium-132 which was precipitated on the hydrogen form of a cation exchange resin saturated with stannous chloride. The daughter iodine was eluted by passing water through the resin. The author claims that the generator gives high yields and requires only simple manipulations. (2)

A German patent claims to separate iodine-132 from tellurium-132 and fission products by precipitating the tellurium and other metal ions with ammonia on a hygroscopic material which adsorbs the ions. The iodine is then rinsed out with a sodium chloride solution, hot or cold water or Ringer's solution. (3)

Several generators which do not rely on adsorption were the early iodine-132 generators.

An early milking system, devised at Brookhaven National Laboratory contained the parent tellurium-132 dissolved in a eutectic mixture of lithium chloride and potassium chloride. In order to milk off the iodine-132 which had grown in, it was necessary first to melt the salt mixture, then sweep out

the iodine by means of an air stream bubbled through the molten liquid. The iodine vapor was adsorbed in a suitable solution. Yields of iodine were good when the vessel containing the molten mixture was glass; however, the mixture rapidly etched the glass, eventually causing it to break. (4)

A second generator, developed at Brookhaven National Laboratory, relies on filtration for the separation of the iodine from precipitated tellurium dioxide. (5) An aqueous solution of sodium hydroxide is added to dissolve the tellurium dioxide which is then reprecipitated by the addition of acetic acid. The iodine-132 which had grown in the solid tellurium dioxide dissolves in the caustic but is not precipitated by the acid. The resulting iodine solution is then filtered off for use.

Another was developed in England by Arnott and Peruma. (6) Their generator was a distillation system in which the parent tellurium was left in contact with a reducing agent, hydroxylamine sulfate. As iodine was required, it was distilled by passing a stream of air through the parent solution. The resulting agitation frees further iodine-132 from the solution. The generator can be recharged merely by adding fresh tellurium to the material already present in the distillation flask. If the yield shows signs of falling below that expected, 0.5 ml

of 20 percent hydrogen peroxide is sucked in. This suppresses the reduction of iodine to iodide by the hydroxylamine.

A similar system is described in a paper by G. B. Cook, J. Eakins and N. Veall. Iodine-132 is allowed to grow in as the iodate; then it is reduced to free iodine immediately before distillation.⁽⁷⁾ A sulfuric acid solution of tellurium-132, containing potassium permanganate, is treated with hydroxylamine sulfate to reduce the daughter iodine-132 from iodate to iodine. The permanganate color is discharged. Some iodine is also reduced to iodide which can be oxidized back to iodine by the addition of a few drops of hydrogen peroxide. The generator is prepared for the next distillation by cooling and adding permanganate, then repeating the process already described.

Practical working generators have been described for milking lanthanum-140 from barium-140, barium-137 from cesium-137, yttrium-90 from strontium-90, iodine-132 from tellurium-132, technetium-99m from molybdenum-99, and gallium-68 from germanium-68.

The first three pairs are separated on Dowex ion-exchange resins. In the first case the parent barium-140 is adsorbed on Dowex 1 x 8 in carbonate form and the daughter lanthanum eluted with either 0.01 M $(\text{NH}_4)_2\text{CO}_3$ or water.⁽⁸⁾ Purity of the product was only fair since it contained about 0.2%

barium-140. Dowex-50 is used by Tracerlab to separate barium-137 from cesium-137, ^(9,10) but no details of the chemical conditions are given. In the yttrium-90 generator, which has been described in detail, ^(11,12) strontium-90 is adsorbed on Dowex-50. The daughter yttrium is eluted with a pH 5.5 citric acid solution. A decontamination factor of 10^{10} is obtainable. The generator is illustrated in Figure 1.

The iodine-132 and technetium-99m generators, originally developed at Brookhaven National Laboratory, are prepared by adsorbing tellurite or molybdate on chromatographic alumina. Elution is accomplished by passing 0.01 M ammonium hydroxide solution through the iodine generator and 0.1 M nitric acid through the technetium generator. Both have been tested thoroughly in the United States and in Europe. ^(11,12, 13,14,15,16,17,18,19) The iodine-132 generator is illustrated in Figure 2. A third generator using alumina was developed at Brookhaven National Laboratory for the milking of gallium-68 from germanium-68 ⁽²⁰⁾. The daughter gallium is eluted rapidly by passing a neutral solution of EDTA (ethylenediamine tetra-acetic acid) through the column. This simple procedure replaces a more inconvenient and time-consuming solvent extraction system. ⁽²¹⁾ Purity of the product is excellent in all three cases and radiocontamination

is less than 10^{-3} % of the product activity. Inactive carrier ions are virtually absent in all three eluates.

Very recently an aluminum-28 generator was developed. (22) It consists of magnesium-28 adsorbed on Dowex-50 cation exchange resin. Aluminum-28 is eluted in ten to thirty seconds, giving a 30% yield, by means of a 1 N sodium hydroxide solution containing 0.5 mg/ml of inactive aluminum. This isotope has the shortest half-life of any nuclide for which a useful generator has been developed.

Other useful generator systems which are either being developed or are under consideration are the pairs yttrium-87 strontium-87m, titanium-44 scandium-44, tin-113 indium-113m, and argon-42 potassium-42. Some desirable characteristics of these systems are the marked differences in length of half life between parent and daughter, and the nature of the daughter radiation. There are many other systems for which workable generators can probably be developed. Undoubtedly, these will receive interest and attention at sometime or other in the future.

A summary of the half-lives and principle radiations of all nuclides discussed above is listed in Table 1.

TABLE 1

Half Lives and Principle Radiations

PARENT				DAUGHTER			
ISOTOPE	$t_{1/2}$	BETA	GAMMA	ISOTOPE	$t_{1/2}$	BETA	GAMMA
Ba ¹⁴⁰	12.8 d	1.02, 0.48	0.54 0.16	La ¹⁴⁰	40.2 h	1.34, 1.10 0.42	1.6, 0.49 0.81, 0.33
Cs ¹³⁷	30 y	0.52, 1.18	-	Ba ^{137m}	2.6 m	I.T.	0.662
Sr ⁹⁰	28 y	0.54	-	Y ⁹⁰	64 h	2.27	-
Te ¹³²	77 h	0.22	0.23	I ¹³²	2.3 h	0.9, 2.12	0.78, 0.67
Mo ⁹⁹	67 h	1.23, 0.45	0.74	Tc ^{99m}	6 h	I.T.	0.142, 0.140
Ge ⁶⁸	250 d	electron capture	X-ray	Ga ⁶⁸	68 m	1.88, 0.78 β^+	1.10
Mg ²⁸	21.3 h	0.45	1.35, 0.95 0.40, 0.032	Al ²⁸	2.3 m	2.87	1.78
Y ⁸⁷	80 h	0.7 β^+	0.48	Sr ^{87m}	2.8 h	I.T.	0.39
Ti ⁴⁴	10 ³ y	electron capture	0.16	Sc ⁴⁴	4.0 h	1.47 β^+	2.54, 1.16
Sn ¹¹³	118 d	electron capture	0.26	In ^{113m}	1.73 h	I.T.	0.39
Ar ⁴²	3.5 y	(3.5, 2.0)	(0.152)	K ⁴²	12.4 h	3.5, 2.0	1.5

FIGURE 1

I^{132} GENERATOR
(ACTUAL SIZE)

FIGURE 2

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