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INTRODUCTION

Promethium-143 has been found useful in studies elucidating the uptake, distribution, excretion and retention in man of the essentially pure beta emitter - ^{147}Pm .⁽¹⁾ Production of ^{143}Pm in these studies was accomplished by proton irradiation of ^{144}Nd ⁽²⁾ and was accompanied by small amounts of ^{144}Pm , the presence of which facilitated deposition measurements by whole-body counting, but which contributed measurably to the dose incurred by the test subjects due to the ^{143}Pm .⁽³⁾ As a consequence of the added dose, a different production method was sought in the hope that ^{143}Pm of greater radionuclidic purity would be produced.

The purpose of this paper is to report the results of a production attempt utilizing proton irradiation of ^{144}Sm and to discuss the dose reduction possible in human studies with the use of the resulting ^{143}Pm .

METHODS AND RESULTS

The production method attempted in the present study primarily involved the reaction:

Other possible, but secondary, reactions with the same target material and proton energy are $^{144}\text{Sm} (p, pn) ^{143}\text{Sm} \longrightarrow ^{143}\text{Pm}$ and $^{144}\text{Sm} (p, 2p) ^{143}\text{Pm}$.

A 174-mg sample of $^{144}\text{Sm}_2\text{O}_3$ (95.1% isotopically enriched) was fabricated into a window-type target⁽⁴⁾ which was irradiated with 22-MeV protons in the 86-inch cyclotron at Oak Ridge National Laboratory for 6.3 hr at an

average beam current of 86.8 μA (integrated beam current = 547 $\mu\text{A-hr}$). The estimated energy of the proton beam at the target was 20.5 ± 0.5 MeV. Following irradiation, the sample was set aside for a short period to allow decay of short-lived activities before processing. The ^{143}Pm product was separated from large amounts of $^{146}\text{-}^{149}\text{Eu}$ activities (formed by (p,n) reactions on isotopic impurities in the original samarium target) and from macro amounts of target material by high-pressure cation exchange chromatography using 0.15 M α -hydroxyisobutyrate (pH = 4.7), AHIB, as the eluting agent. The carrier-free ^{143}Pm product was co-precipitated with $\text{Fe}(\text{OH})_3$ to separate it from the AHIB and then separated from iron by anion exchange chromatography in 6 M HCl solution.⁽⁵⁾

It is estimated that the product solution contained approximately 257 μCi of ^{143}Pm so that, for this irradiation configuration, the production rate and cyclotron yield of ^{143}Pm was 0.47 $\mu\text{Ci}/\mu\text{A-hr}$ and 41 $\mu\text{Ci/hr}$, respectively.

A chemical spot test for iron indicated <5 μg of Fe^{3+}/ml in the final product solution. The samarium content of the solution was <50 ppb as measured with a spark source mass spectrograph indicating a very clean separation of the ^{143}Pm from the target material. Gamma energy analysis of the product solution indicated a ^{143}Pm activity of 210 μCi resulting in an estimated recovery of ^{143}Pm for the above-described chemical separation of approximately 80%.

The photon spectrum of the product solution as measured with a lithium-drifted germanium detector is shown in Figure 1. The photopeaks observed can be attributed to ^{143}Pm only with no evidence of gamma-emitting contaminants being present with energies up to 2 MeV. Analysis

of the product solution for alpha contaminants by 2π alpha proportional counting was negative, i.e., no alpha activity was detected above the background of the instrument which, for the measurement conditions, corresponded to an alpha disintegration rate of <4 d/m.

DISCUSSION

The ^{143}Pm used in previous human metabolism studies was produced by the reaction, $^{144}\text{Nd} (p,2n) ^{143}\text{Pm}$, in which enriched $^{144}\text{Nd}_2\text{O}_3$ (94.4%) was used as the target material. Two such production attempts were made and a summary of production parameters for each run is presented in Table 1. Similar data for ^{143}Pm production by proton irradiation of ^{144}Sm are also presented in Table 1 for comparison. The production data for both methods indicate that although the ^{143}Pm yield of the $^{144}\text{Nd} (p,2n)$ reaction is some 3 to 4 times higher than that of the $^{144}\text{Sm} (p,2n)$ reaction, the production of ^{144}Pm is also considerably higher in the former reaction. Approximately 5% of the total promethium activity produced in the $^{144}\text{Nd} (p,2n)$ reaction is due to ^{144}Pm which is produced in a competing (p,n) reaction. Since the half-life of ^{144}Pm is longer than that of ^{143}Pm (349 days and 267 days, respectively⁽⁶⁾) the relative activity of ^{144}Pm in a $^{143}\text{-}^{144}\text{Pm}$ mixture will increase with time after production. The extent of this increase is shown in Figure 2 where it is seen that a $^{143}\text{-}^{144}\text{Pm}$ mixture containing 5.5% by activity ^{144}Pm at production will decay to about 40% of its initial activity after one year, but its ^{144}Pm activity will have increased to almost 7% of the total activity. At the end of three years, the ^{144}Pm activity will be about 10% of the total activity.

This increase is particularly significant in terms of the dose

incurred by volunteers who are administered ^{143}Pm for human metabolism studies. Table 2 presents a comparison of the calculated doses based on standard man parameters⁽⁷⁾ to selected critical organs of volunteers administered ^{143}Pm and ^{144}Pm as the chloride by intravenous injection and ingestion. Based on unit activity, the doses from ^{144}Pm to the organs listed are some 4 to 5 times higher than those from ^{143}Pm .

Table 3 presents the calculated doses to the same critical organs per μCi ^{143}Pm containing 5%, 7.5% and 10% by activity of ^{144}Pm . A comparison of the data in Tables 2 and 3 indicates that, per unit activity of ^{143}Pm , the doses to the critical organs will be increased by an average of about 21% with material that contains 5% ^{144}Pm by activity. Similarly, administered promethium containing 7.5% and 10% ^{144}Pm by activity will result in an estimated 32% and 44% higher dose, respectively, than that resulting from the same activity of promethium administered as ^{143}Pm .

In the previous studies involving administration of promethium to humans, levels of 0.1 μCi ^{143}Pm for intravenous injection and 10 μCi for ingestion were chosen as a balance between enough activity to follow the body burdens for about one year and the smallest possible doses to the volunteers.⁽¹⁾ The resulting doses were extremely small in light of the fact that for the highest exposed organ, the lower large intestine, the one time test subject dose was still less than one percent of the maximum permissible annual limit. But in keeping with the stated philosophy of the ICRP⁽⁷⁾ that "all doses be kept as low as practicable and that any unnecessary exposure be avoided", the use of ^{143}Pm uncontaminated with ^{144}Pm seems to be indicated in order to maintain the dose to volunteer test subjects at an insignificant level.

CONCLUSIONS

Promethium-143 of high radionuclidic purity can be produced by proton irradiation of ^{144}Sm . The use of such ^{143}Pm in future human metabolism studies as a substitute for ^{147}Pm insures that doses to volunteer test subjects will be reduced by about 25% compared to the doses received in previous studies in which ^{143}Pm containing small amounts of ^{144}Pm was used.

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TABLE 1: Summary of ^{143}Pm Production Parameters
for Proton Irradiation of ^{144}Nd and ^{144}Sm

Production Method	$^{144}\text{Nd} (p,2n) ^{143}\text{Pm}$	$^{144}\text{Sm} (p,2n) ^{143}\text{Eu}$ → ^{143}Sm → ^{143}Pm	
Irradiation Date	2/3/66	7/25/67	2/25/70
Proton Energy at Target (MeV)	20.5±0.5	20.5±0.5	20.5±0.5
Average Beam Current (μA)	80.7	86.8	86.8
Irradiation Time (hr)	1.0	1.0	6.3
Integrated Beam Current (μA-hr)	80.7	86.8	547
^{143}Pm Produced (μCi)	115	154	257
^{143}Pm Production Rate (μCi/μA-hr)	1.4	1.8	0.47
^{143}Pm Cyclotron Yield (μCi/hr)	115	154	41
^{144}Pm Produced (% of Total Pm Activity)	5.1	5.5	0.07

TABLE 2: Calculated Doses to Critical Organs
 Following I.V. Injection and Ingestion of ^{143}Pm and ^{144}Pm

Intravenous Injection

<u>Critical Organ</u>	<u>$^{143}\text{PmCl}_3$</u>	<u>$^{144}\text{PmCl}_3$</u>
Total Body	50 mrem/ μCi	260 mrem/ μCi
Bone	100 "	410 "
Liver	400 "	1930 "

Ingestion

<u>Critical Organ</u>	<u>$^{143}\text{PmCl}_3$</u>	<u>$^{144}\text{PmCl}_3$</u>
Stomach	0.52 mrem/ μCi	1.9 mrem/ μCi
Small Intestine	0.97 "	3.8 "
Upper Large Intestine	4.5 "	16 "
Lower Large Intestine	9.2 "	32 "
Total Body	0.005 "	0.026 "
Bone	0.01 "	0.041 "
Liver	0.006 "	0.029 "

TABLE 3: Estimated Doses to Critical Organs
per μCi of ^{143}Pm Containing Various Levels of ^{144}Pm

Intravenous Injection

<u>Critical Organ</u>	<u>5% ^{144}Pm</u>	<u>7.5% ^{144}Pm</u>	<u>10% ^{144}Pm</u>
Total Body	63	69	76
Bone	120	130	140
Liver	500	550	590

Ingestion

<u>Critical Organ</u>	<u>5% ^{144}Pm</u>	<u>7.5% ^{144}Pm</u>	<u>10% ^{144}Pm</u>
Stomach	0.62	0.66	0.71
Small Intestine	1.2	1.3	1.4
Upper Large Intestine	5.3	5.7	6.1
Lower Large Intestine	10.8	11.6	12.4
Total Body	0.006	0.007	0.008
Bone	0.012	0.013	0.014
Liver	0.007	0.008	0.009

FIGURE 1
Ge(Li) DETECTOR SPECTRUM OF ¹⁴³Pm PRODUCED BY PROTON IRRADIATION OF SAMARIUM

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FIGURE 2
 VARIATION WITH TIME OF TOTAL ACTIVITY AND ^{144}Pm CONTENT OF PROMETHIUM PRODUCED
 BY PROTON IRRADIATION OF NEODYMIUM

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