jnm / CONCISE COMMUNICATION

DETERMINATION OF IMPURITY ACTIVITIES IN FISSION-PRODUCT GENERATOR ELUATE

D. W. Anderson, D. E. Raeside, and V. J. Ficken

University of Oklahoma Health Sciences Center, Oklahoma City, Oklahoma

Impurity activities in the eluate from fission-product technetium generators have been studied by taking pulse-height spectra with a Ge(Li) spectrometer. The impurities ^{99}Mo, ^{131}I, ^{132}I, and ^{133}I have been found in the eluate with activities in the microcurie and submicrocurie range. The measured contaminant activities per millicurie of ^{99m}Tc were variable but within safe limits except in a special circumstance when the ^{131}I activity was excessively large.

The subject of impurities in ^{99m}Tc eluate used for nuclear medicine procedures has been discussed in several articles in the last few years (*1–4*). Since the United States Atomic Energy Commission has limited the level of the contaminant activity that can be administered to patients by holders of by-product licenses, users are required to perform radionuclide impurity checks on a routine basis, particularly for ^{99}Mo. In recent years the demand for ^{99}Mo–^{99m}Tc generators yielding high concentrations of ^{99m}Tc has resulted in the manufacture of generators using fission-produced ^{99}Mo. The contaminants in eluate from these generators differ from the contaminants observed in the generators made from neutron-irradiated molybdenum, since the yield of radioisotopes from any nuclear reaction is specific to the reaction type and target involved.

In a previous paper (*5*) the results of some measurements made with a NaI(Tl) detector system and a multichannel pulse-height analyzer were reported. Some of the difficulties which occur when a scintillation detector is used to monitor eluate from a fission-product generator were discussed. In particular, because of the relatively poor resolution of the system and because the contaminant radioisotopes have complex decay schemes, overlapping spectrum peaks from different radioisotopes are likely to appear on NaI(Tl) spectra. Recognizing these difficulties Briner and Harris (*6*) have suggested some techniques using scintillation detectors, which facilitate identification of ^{99}Mo or ^{132}I activities when one is dominant. When both impurities are present in substantial quantities, a stripping technique using spectra from a multichannel analyzer was shown (*6*) which is applicable for quantitative determinations. The disadvantage in this technique is that considerable time must elapse for decay of the 2.3-hr ^{132}I before the procedure can be completed.

In the following communication we describe measurements made with a high-resolution Ge(Li) spectrometer that are quick and precise. In addition the technique used is appropriate for detection of any radioactive impurity which emits gamma rays.

EQUIPMENT AND PROCEDURES

Daily monitoring for contaminant activities in the eluate from fission generators purchased from a single supplier was carried out for a period in excess of 6 months with a NaI(Tl) detection system with lead absorber and 256-channel pulse-height analyzer. A pulse-height spectrum was obtained routinely after each elution. If the spectrum showed a single impurity photopeak at 740–780 keV, the ^{99}Mo activity was computed by the method discussed in Ref. *5*. If several large photopeaks were apparent or unusually broad and asymmetric peaks were seen, or if the photopeak energy was seemingly shifted, generator eluate was subsequently studied with a Ge(Li) spectrometer.

A brief description of the spectrometer is given below. For a complete discussion of Ge(Li) detector systems and operations see Refs. *7* and *8*. In our system a 30-cm^3 lithium-drifted germanium detector, Ge(Li), with about 3-keV resolution (at 1333 keV)

Received Feb. 11, 1974; revision accepted April 29, 1974.

For reprints contact: David W. Anderson, Radiological Sciences, University of Oklahoma Health Sciences Center, P.O. Box 26901, Oklahoma City, Okla. 73190.

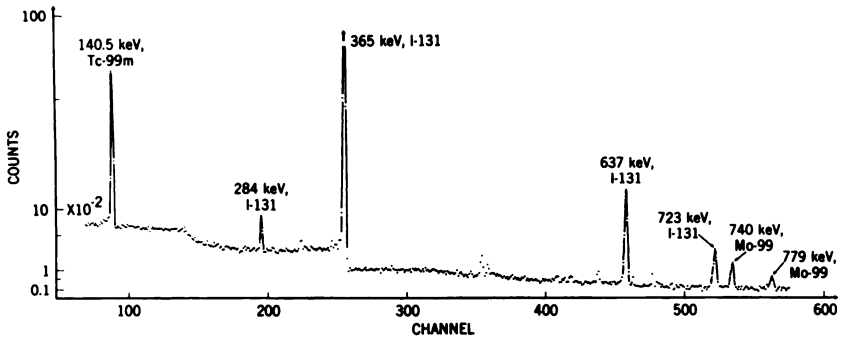

FIG. 1. An eluate pulse-height spectrum with ^{131}I gamma-ray peaks is shown. The $^{131}I/^{99m}Tc$-activity ratio was 0.05 μCi/mCi at time of elution.

was used with a 1024-channel pulse-height analyzer. Since the eluate contained large amounts of ^{99m}Tc, a 4-mm-thick piece of lead was used between the detector and the eluate bottle as a filter for the 140-keV gamma rays. To specify gamma-ray energies and make quantitative evaluations, the system had to be calibrated for energy and for absolute efficiency. These measurements were made with a mixed radionuclide standard calibrated for emission rates by the National Bureau of Standards. The solution contained ^{109}Cd (88 keV), ^{57}Co (122 keV), ^{109}Ce (166 keV), ^{203}Hg (279 keV), ^{113}Sn-^{113m}In (392 keV), ^{85}Sr (514 keV), ^{137}Cs (662 keV), ^{88}Y (898, 1836 keV)* and ^{60}Co (1173, 1333 keV). Since the emission standard was in a glass bottle filled with about 50 ml of solution, all eluate samples were diluted to the same volume and counted in similar bottles.

RESULTS AND DISCUSSION

The principal contaminant activities found in the eluate were ^{99}Mo, ^{131}I, ^{132}I, and ^{133}I. Traces of ^{103}Ru and possibly ^{105}Rh have also been identified. Figures 1 through 3 show pulse-height spectra obtained with the Ge(Li) spectrometer and allow identification of the principal radioactive impurities. Notice that the data are plotted on square-root graph paper (*9,10*). This type of plot allows a large range of peak counts and guarantees that the scatter of points due to Poisson fluctuations is independent of the position of the points on the plot. Since the spectra were taken through a lead filter, the 140-keV ^{99m}Tc peak is greatly suppressed. Lead x-rays and the backscatter peak for the 140-keV ^{99m}Tc gamma rays dominate at energies below 100 keV so this region is not shown. Data for the figures were taken several hours after elution and the impurity activities which were identified ranged from 0.1 to 10 μCi.

* Because ^{88}Y decays by β^+ emission the 511-keV gamma ray will be present in the standard spectrum. This means that the 514-keV ^{85}Sr peak is not suitable for precise detector efficiency measurements.

Notice that ^{131}I has been identified by several of the characteristic gamma rays, not merely by a peak at 365 keV. Since ^{99}Mo decays with emission of a 366-keV gamma ray with about 1% probability (*11*), samples with high ^{99}Mo activity could cause erroneous identification of ^{131}I.

A brief discussion of the production of iodine in fission generators is in order. Iodine-132 has a relatively short half-life (2.3 hr) and therefore is present in the eluate as the ^{132}Te daughter. The ^{132}Te parent has a 78-hr half-life so the equilibrium ^{132}I activity must decrease in time with this half life. Nevertheless, the ratio of ^{132}I activity to ^{99m}Tc activity in the column increases with the passage of time since ^{132}Te has a longer half-life than ^{99}Mo, the ^{99m}Tc parent. Thus ^{132}I contamination presents more of a problem near the end of the useful lifetime of a generator than at the beginning.

Iodine-131 and ^{133}I are produced copiously in ^{235}U fission and because of their long half-lives would be expected as impurities. In addition, some ^{131}I is produced as the daughter of ^{131m}Te (30 hr). Of these two, the ^{131}I causes the most difficult contamination problem because of its very long half-life when compared with ^{99m}Tc and even ^{99}Mo. Thus the ^{131}I activity per millicurie of ^{99m}Tc is likely to be longest near the end of the useful lifetime of the generator (assuming that only a small percentage of the ^{131}I washes out of the column in any single elution). In addition the ratio of ^{131}I activity to ^{99m}Tc activity increases as the time between elution and administration to the patient increases.

AEC radiopharmaceutical criteria (*12*) specify that the maximum allowed impurity levels present at the time of administration to the patient are as follows: 1 μCi of ^{99}Mo/mCi of ^{99m}Tc, 0.05 μCi of ^{131}I and ^{103}Ru/mCi of ^{99m}Tc, and 0.1 μCi/mCi of ^{99m}Tc for all other activities. Although our work is in no way an exhaustive study of impurities in the eluate from these generators, our experience has been that impurity-to-^{99m}Tc-activity ratios are usually more than a factor of 10 below these levels. Occasionally the contaminant yield has been sub-

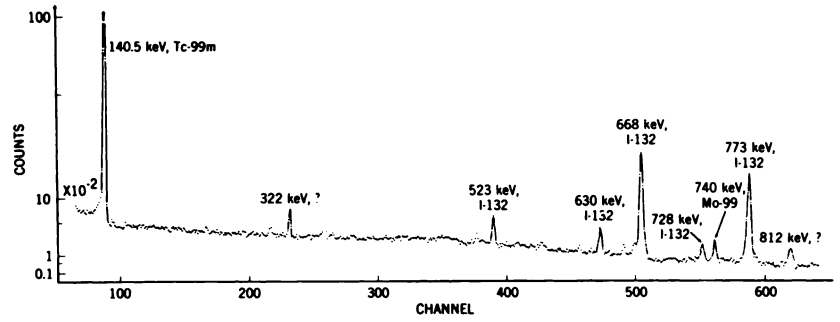

FIG. 2. An eluate pulse-height spectrum with ^{132}I gamma peaks is shown. The ^{132}I/^{99m}Tc-activity ratio was 0.01 μCi/mCi at time of elution.

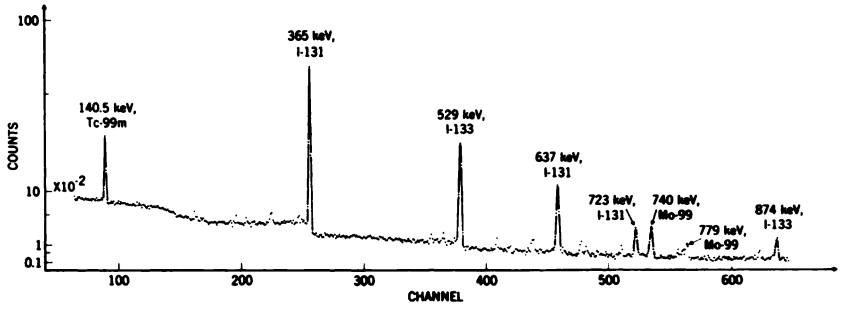

FIG. 3. An eluate pulse-height spectrum with ^{131}I and ^{133}I gamma peaks is shown. The ^{131}I/^{99m}Tc-activity ratio was 0.01 μCi/mCi at time of elution. The ^{133}I-to-^{99m}Tc-activity ratio was 0.002 μCi/mCi at time of elution.

stantially larger than normal. The spectra and activity ratios given in Figs. 1 through 3 show some of the largest contaminant yields. In one case for special circumstances the impurity/^{99m}Tc-activity ratio was found to exceed the specifications. This was for ^{131}I and the large activity ratio was measured when the generator was eluted the second week after delivery and when the eluate was assayed several hours after the elution.

CONCLUSIONS

Although high-yield fission product generators are especially useful in many clinical situations, the iodine impurity is of some concern. Because of this we recommend that the eluate from such generators be monitored with a high-resolution spectrometer. Unfortunately, because this type of spectrometer is expensive and not commonly available, such a procedure may not be possible in most user laboratories. As suggested in previous work (*5*), the problem would be considerably alleviated if high-resolution spectrum information were routinely furnished by the supplier with each fission-product generator. This would be convincing documentation of the purity of the radioisotope yield. In any case ^{131}I contamination should be evaluated especially when the fission-generator ^{99m}Tc yield is small.

ACKNOWLEDGMENTS

We wish to thank the Department of Aerospace, Mechanical, and Nuclear Engineering of the University of Oklahoma for the use of their equipment and John L. James in particular for his technical assistance.

REFERENCES

1. SMITH EM: Properties, uses, radiochemical purity and calibration of ^{99m}Tc. *J Nucl Med* 5: 871–882, 1964

2. CROSBY EH: Radiochemical purity of short-lived technetium 99m from commercial suppliers. *Radiology* 93: 435–439, 1969

3. ROBISON GD: Impurities in ^{99m}Tc-Sodium pertechnetate produced by methyl-ethyl-ketone extraction. *J Nucl Med* 13: 318–320, 1972

4. BARDFIELD PA, RUDIN S: Comparative study of instant technetium from various commercial suppliers. *J Nucl Med* 14: 880–881, 1973

5. FICKEN V, ANDERSON D, COX P, et al: Radionuclide contamination of fission generator eluate. *J Nucl Med Technol* 1: 11–13, 1973

6. BRINER WH, HARRIS CC: Radionuclide contamination of eluates from fission-product molybdenum-technetium generators. *J Nucl Med* 15: 466–467, 1974

7. HOFFER PB, BECK RN, GOTTSCHALK A: The role of semiconductor detectors in the future of nuclear medicine. New York, Society of Nuclear Med, Inc, 1971, pp 1–37

8. FIEDLER H, TENCH O: Germanium (Li) gamma spectrometer systems. Canberra Industries Inc, 1968

9. HOOTEN IN: The display of statistical data from nuclear counting experiments. *Nucl Instr Meth* 56: 277–283, 1967

10. VERRALL RI, BELL RE: Square root graph paper for nuclear spectra. *Nucl Instr Meth* 67: 353–354, 1969

11. DILLMAN LT: Radionuclide decay schemes and nuclear parameters for use in radiation-dose estimation. MIRD Pamphlet No 6, *J Nucl Med* 11: Suppl No 4, 7–32, 1970

12. Radiopharmaceutical criteria for ^{99}Mo-^{99m}Tc generators containing fission produced parent ^{99}Mo. USAEC, 15 Nov, 1971