# Radionuclide Contamination of Fission Generator Eluate

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The need for $^{99m}$Tc generators producing high yields in a small eluate volume has led to the use of fission-produced $^{99}$Mo and generator column changes necessary to accommodate much smaller quantities of molybdenum. Because of these changes, the Atomic Energy Commission distributed new specifications for maximum radionuclide contamination (*1*) as early as November 15, 1971. Our laboratory has encountered contamination problems involving commercially available fission generators which we believe warrant discussion.

Several methods of monitoring microcurie quantities of $^{99}$Mo in the presence of hundreds of millicuries of $^{99m}$Tc have been used with apparent success. In the following we will compare data obtained using a recommended lead filtration technique* and a single-channel analyzer with data resulting from a multichannel analyzer technique. This information was obtained for small eluate volumes and high $^{99m}$Tc activity levels. In addition, we will consider contaminants other than $^{99}$Mo which can cause difficulty in the $^{99}$Mo determination.

## Determination of $^{99}$Mo Contamination

To filter out most of the $^{99m}$Tc photons, a 5-mm-thick lead disk was used. Then a direct comparison of the transmitted portion of the eluate photon flux with a $^{99}$Mo standard source filtered in the same way could be made. During the procedure the 5-ml generator eluate and 5-ml $^{99}$Mo standard were each contained in 30-ml eluate vials which were shielded by identical lead containers with a 1.5-cm-diam port.

Following energy calibration of each detector-analyzer system with a $^{137}$Cs source, both eluate and $^{99}$Mo standard were counted under identical geometric conditions. Photon energies between 600 and 900 keV were counted with a single-channel analyzer (SCA) while photon energies above 10 keV were collected with the multichannel analyzer (MCA). Conversion of $^{99}$Mo counts per 5 min from the standard yielded a factor of $3.37 \times 10^{-3}$ $\mu$Ci of $^{99}$Mo per count from the SCA. By adding counts per channel for a 200-sec live-time spectrum from the MCA a factor of $3.9 \times 10^{-4}$ $\mu$Ci of $^{99}$Mo per count was obtained. In this case the interval was from the valley on the low-energy side through the 740—780-keV $^{99}$Mo peak to the background level on the high-energy side. Room background did not significantly contribute to the counts. These factors were then multiplied by eluate counts on a day-to-day basis to yield activity of $^{99}$Mo in microcuries. Comparative results selected at random are listed in Table 1. The data agree reasonably well.

With the use of the MCA, an additional correction may be made which could not be readily achieved with the SCA data. Figure 1A is the spectrum of a $^{99}$Mo standard after lead filtration obtained by the MCA. If it were possible to subtract only the 740—780-keV events which are in the major peak, a spectrum similar to that diagrammed in Fig. 1B might be anticipated. The counts in the shaded area are there as a result of summing

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**Table 1. Paired Results of $^{99}$Mo Assays from SCA and MCA**

| SCA $\mu$Ci $^{99}$Mo | MCA $\mu$Ci $^{99}$Mo | MCA $\mu$Ci $^{99}$Mo with summing correction |
|---|---|---|
| 7.3 | 7.3 | 7.3 |
| 8.6 | 8.9 | 9.1 |
| 3.9 | 4.1 | 3.5 |
| 2.4 | 2.7 | 2.8 |
| 3.9 | 3.7 | 3.6 |
| 21.5 | 23.8 | 15.6 |
| 14.0 | 12.2 | 12.2 |
| 5.6 | 7.8 | 6.4 |

**MolyTech Calibrator.* Registered by Mallinckrodt Nuclear. St Louis, Mo

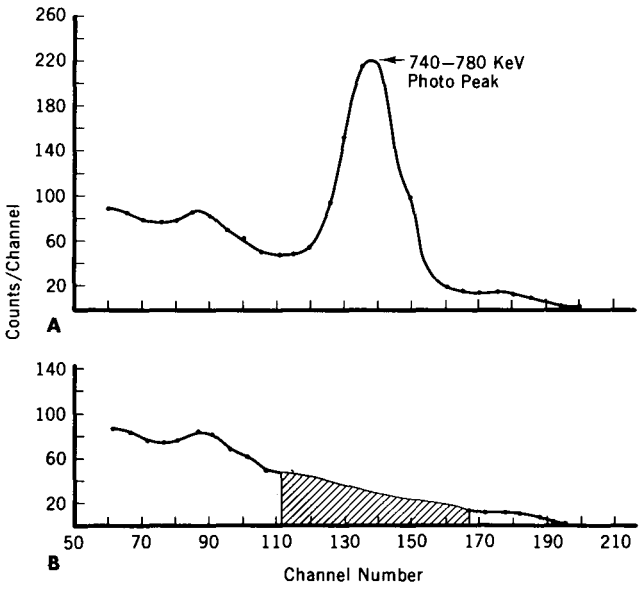


**FIG. 1.** (A) $^{99}$Mo spectrum after lead filtration. (B) $^{99}$Mo spectrum after lead filtration with 740–780-keV contribution subtracted (diagram).

$^{99m}$Tc with other contaminants. By subtracting the counts in each channel of the shaded area of Fig. 1B from each corresponding channel in Fig. 1A, a correction for summing is made. This method provided a ratio of $6.04 \times 10^{-4}$ $\mu$Ci of $^{99}$Mo per count. When this method is applied and the actual number of counts contributed by the $^{99}$Mo in the unknown is multiplied by the ratio, microcuries of $^{99}$Mo per total eluate are obtained (Table 1). If sufficient lead shielding is used to filter out the $^{99m}$Tc, summing corrections will be negligible. It should be noted that one correction resulted in a significant change and was due to the presence of a contaminant with a gamma energy just below that of $^{99}$Mo.

## Other Radionuclide Contaminants

We have demonstrated that for routine analysis with only $^{99}$Mo contamination, SCA-lead filter techniques yield nearly the same assay results as more elaborate MCA-lead filter techniques. However, complications frequently arise which could render the SCA methods invalid. Incomplete separation of $^{99}$Mo from other fission products can result in gamma peaks in the vicinity of the $^{99}$Mo peak and would render the $^{99}$Mo assay abnormally high. In addition, contaminant photopeaks outside the typical 600–900-keV window would be totally unobserved by the user. We have observed both situations.

Figure 2A is a spectrum of $^{99}$Mo standard after 5-mm lead filtration. Figure 2B is the spectrum after lead filtration from the eluate of a commercial fission generator. Our scintillation detector system could not resolve the two peaks of $^{99}$Mo at 740–780 keV (Fig. 2A); however, the broad appearance of the eluate spectrum did reveal the presence of another radionuclide with similar gamma energy (Fig. 2B). The gamma energies were identified with the use of a high-resolution Ge(Li) detector as being 519 ± 2 keV, 668 ± 2 keV, and 773 ± 2 keV, representing $^{132}$I decay. Since the $^{132}$I gammas contribute to what is normally the $^{99}$Mo peak at 740–780 keV, determination of $^{99}$Mo with a SCA would be invalid. At best one could only make a good guess with a NaI(Tl) MCA system. Moreover, the contamination of the $^{132}$I could in principle exceed the AEC specifications.

Figure 3 is a spectrum after lead filtration from the eluate of another generator from the same supplier. At least three significant peaks other than the 740–780-keV peak of $^{99}$Mo have been identified. The gamma energies are 364 ± 2 keV, 497 ± 2 keV, and 530 ± 2 keV. These energies were obtained by the use of the Ge(Li) detector. These contaminants have been tentatively identified as $^{131}$I, $^{103}$Rb, and $^{133}$I. Obviously the use of a SCA and a window observing photon energies between 600 and

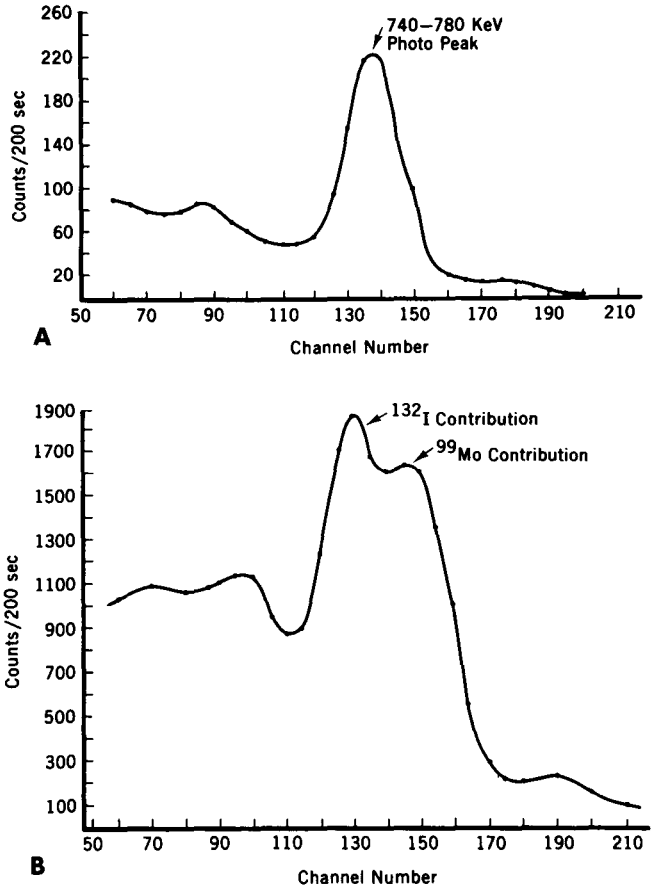


**FIG. 2.** (A) $^{99}$Mo spectrum after lead filtration. (B) Fission generator eluate spectrum after lead filtration.

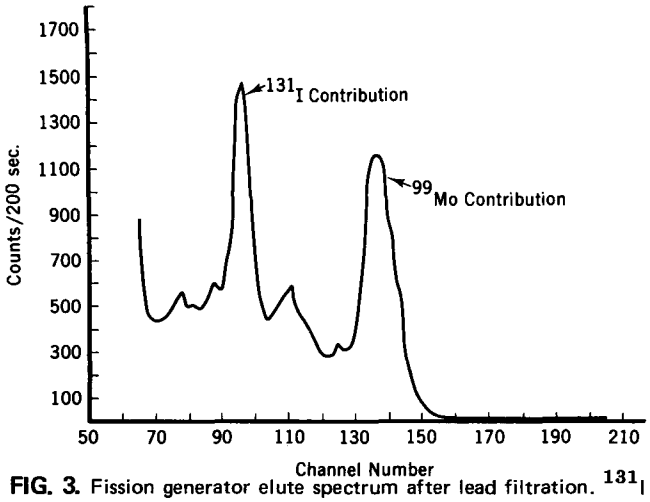


**FIG. 3.** Fission generator elute spectrum after lead filtration. $^{131}I$ photopeak is clearly present.

900 keV would preclude the detection of any gamma energies below this level. Unfortunately even the use of a multichannel analyzer scintillation detector system to quantitate the activity is a time-consuming task.

## Conclusions

Under the present circumstances it would appear that the eluate from fission generators should be monitored by a MCA detector system to be certain of radionuclide purity. Unfortunately even multichannel analysis with a scintillation detector may not be sufficient to allow accurate measurement of the activities of each of the contaminants which are frequently present. A cooled Ge(Li) detector may be required to identify these contaminants.

Not every nuclear medicine laboratory which may require a fission generator can afford such elaborate instrumentation. In view of this, it would be useful for commercial suppliers to obtain detailed gamma analysis of the first eluate from each generator before it is shipped and provide the user with high-resolution spectrum information. Since $^{99}Mo$ contamination in the eluate is always present and could usually be monitored by the user, the efficiency of the user's SCA system would be known for $^{99}Mo$ after lead filtration. The supplier could then provide the user a factor based on decay modes, differential lead absorption of the contaminant gamma energies, and window settings. This factor, when applied to the activity of $^{99}Mo$ determined by the user and counts from the contaminant photopeak, would yield approximate levels of that radionuclide impurity. This would be an improvement over present procedures, although admittedly a compromise.

## References

1. *Radiopharmaceutical Criteria for $^{99}Mo/^{99m}Tc$ Generators Containing Fission Produced Parent $^{99}Mo$.* USAEC, 15 Nov, 1971