

## Wavelengths of the M X-Ray Spectra of Uranium, Neptunium, Plutonium, and Americium \*

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The M series of the X-ray spectra of uranium, neptunium, plutonium, and americium have been measured between 250 and 520 pm by means of a linear spectrometer. The relative intensities of the lines have been normalized to the respective  $M\alpha_1$  lines of the actinides.

The qualitative and quantitative analysis of the actinides is becoming increasingly important for the characterization of nuclear fuels in the as-fabricated conditions and after irradiation as well as of residues from the reprocessing of burnt fuels. The identification of these elements requires the precise knowledge of the wavelengths and the intensities of the X-ray emission spectra of the M series which have a favourable wavelength region between 200 and 500 pm for the electronprobe microanalysis. X-ray emission spectra of the M lines of thorium, protactinium, and uranium have been reviewed [1]; further measurements on uranium have also been reported [2, 3, 10]. The major M lines of the transuranium elements have been analyzed recently in most cases on metallic specimens of neptunium [4, 5, 6], plutonium [5, 7, 8, 9, 10], and americium [5].

In this study the complete series of the M lines of uranium, neptunium, plutonium, and americium have been measured between 250 and 520 pm. Sintered  $UO_2$ , 2%  $NpO_2$  dissolved in a borosilicate glass, sintered  $PuO_2$ , and a two-phase  $AmAl_4$ -Al alloy have been prepared by the hot cell metallographic techniques. The samples were embedded into araldite resin and were polished with 0.25  $\mu m$  diamond paste. The homogeneity was checked by  $\alpha$  autoradiography.

The instrument consists of an  $\alpha$  and  $\gamma$  shielded electronprobe microanalyzer JRXA 50 (Japan

Electron Optics Ltd.) which is connected to a shielded cell. The sample loading was carried out inside of the cell from which an automatic and radiation protected transport into the rear of the instrument is guaranteed. The spectra have been analyzed using a linear spectrometer in Johann geometry with a Rowland radius  $R = 180$  mm and a constant take-off angle of  $35^\circ$  between the polished sample surface and the X-ray which is diffracted on the 1011 plane of a quartz crystal ( $2d = 668.62$  pm). The distance  $b$  between sample and crystal is not constant but varies linearly with wavelength  $\lambda$  and  $\sin \theta$ , resp. The spectra have been recorded continuously with a proportional counter in cps using 20 kV electron accelerating voltage and 100 to 250 nA sample current (Figure 1). The high precision analysis has been made by count rate measurements by varying  $b$  in steps of 0.05 mm which are approximately equivalent to 0.093 pm. The wavelength measurements were absolute using the relation  $\lambda = b \cdot d/R$  and were calibrated for cross-check using  $K\alpha_1$  and  $L\alpha_1$  lines of internal standards, resp. The error in the wavelength measurements is  $\pm 0.1$  pm for the stronger lines; it is  $\pm 0.2$  pm for intensities lower than 3% of the strongest line of the respective M spectrum.

The measured wavelengths and the relative intensities ( $M\alpha_1 = 100\%$ ) of the M series of uranium, neptunium, plutonium, and americium are given in Table 1. The  $M\alpha_2$  lines could be detected on the

Table 1. Wavelengths  $\lambda$  (in pm) and relative intensities r. i. ( $M\alpha_1 = 100\%$ ) of the M series of the X-ray lines.

| line                        | uranium   |      | neptunium |      | plutonium |      | americium |      |
|-----------------------------|-----------|------|-----------|------|-----------|------|-----------|------|
|                             | $\lambda$ | r.i. | $\lambda$ | r.i. | $\lambda$ | r.i. | $\lambda$ | r.i. |
| $M_I N_{III}$               | 275.4     | 1    | —         | <1   | 258.1     | <1   | —         | <1   |
| $M_{II} N_{IV}$             | 281.9     | 2    | 272.3     | 1    | 264.1     | 1    | 255.5     | 3    |
| $M_{II} N_{II}$             | 290.7     | 1    | —         | <1   | —         | <1   | —         | <1   |
| $M_{III} O_{IV,V}$          | 295.1     | 5    | 286.2     | 2    | 278.3     | 3    | 270.4     | 3    |
| $M_{II} N_I$                | 333.1     | 2    | 322.2     | 2    | 312.7     | 3    | 303.0     | 6    |
| $M_{III} N_V (\zeta_1)$     | 347.9     | 13   | 338.7     | 3    | 329.5     | 6    | 320.6     | 7    |
| $M_{III} N_{IV} (\zeta_2)$  | 352.0     | 2    | 342.5     | <1   | 333.7     | 1    | 324.9     | 1    |
| $M_{IV} O_{II}$             | 357.6     | 1    | —         | <1   | 336.2     | <1   | —         | <1   |
| $M_{IV} N_{VI} (\beta)$     | 371.6     | 180  | 360.8     | 72   | 351.0     | 67   | 341.3     | 64   |
| $M_{IV} N_{VII} (\alpha_1)$ | 391.0     | 100  | 380.0     | 100  | 370.1     | 100  | 360.2     | 100  |
| $M_{III} N_I$               | 433.0     | 0.5  | 421.5     | 0.2  | 412.2     | 0.2  | 402.2     | 0.2  |
| $M_V N_{III} (\zeta_1)$     | 494.3     | 0.9  | 480.4     | 0.3  | 467.2     | 0.2  | 454.3     | 0.2  |
| $M_V N_{II} (\zeta_2)$      | 504.9     | 0.6  | 491.3     | 0.2  | 478.4     | 0.2  | 466.2     | 0.2  |

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high wavelength tails of the  $M\alpha_1$  lines for all four elements investigated. However, they were too weak in intensity for a precise wavelength determination. The  $M\zeta$  lines of neptunium, plutonium, and americium and the  $M_{III}N_{IV}$  and  $M_{III}N_I$  transitions of neptunium and americium have been measured for the first time. The results of the

remaining lines of uranium, plutonium, and americium agree very well with the literature values. However, the wavelengths of neptunium are throughout 0.1 to 0.5 pm lower than the values cited in literature in spite of the fact that different internal standards (Ce  $L\alpha_1$ , Ti  $K\alpha_1$ , Ca  $K\alpha_1$ ) were used for the analysis of this element.

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