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# Criticality of a $^{237}$Np Sphere

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A critical mass experiment using a 6-kg $^{237}$Np sphere has been performed. The purpose of the experiment is to get a better estimate of the critical mass of $^{237}$Np. To attain criticality, the $^{237}$Np sphere was surrounded with 93 wt % $^{235}$U shells. A 1/M as a function of uranium mass was performed. An MCNP neutron transport code was used to model the experiment. The MCNP code yielded a $k_{eff}$ of 0.99089 ± 0.0003 compared with a $k_{eff}$ 1.0026 for the experiment. Based on these results, it is estimated that the critical mass of $^{237}$Np ranges from kilogram weights in the high fifties to low sixties.

***KEYWORDS: neptunium, critical mass, fast spectrum, bare, metal, sphere, highly enriched uranium***

## 1. Introduction

For the past five years, scientists at Los Alamos National Laboratory have mounted an unprecedented effort to get a better estimate of the critical mass of $^{237}$Np. To accomplish this task, a 6-kg $^{237}$Np sphere was recently cast[1] and used in this experiment.

It is well known that $^{237}$Np is primarily produced by successive neutron capture events in $^{235}$U or through the (n, 2n) reaction in $^{238}$U. These nuclear reactions (see Eq. 1) lead to the production of $^{237}$U, which decays by beta emission into $^{237}$Np.

$$^{235}\mathrm{U}(n,\gamma)\,^{236}\mathrm{U}$$

$$^{236}\mathrm{U}\,(n,\gamma)\,^{237}\mathrm{U} \rightarrow \beta \rightarrow {}^{237}\mathrm{Np} \qquad (1)$$

$$^{238}\mathrm{U}\,(n, 2n)\,^{237}\mathrm{U} \rightarrow \beta \rightarrow {}^{237}\mathrm{Np}$$

It is estimated that a typical 1000 MW (e) reactor produces on the order of 13 kg of neptunium in a year.[2] Some of the neptunium in irradiated fuel elements from production reactors has been separated and is presently stored in containers in a liquid form. This form or method of storage is adequate from the point of view of criticality safety because the fission cross section for $^{237}$Np at thermal energies is quite low and any moderation of the neutron population would increase the critical mass of $^{237}$Np to infinity. However, for long-term storage, the neptunium liquid solutions must be converted to oxide and metal because these forms are less movable and less likely to leak out of containers. At the present, there is some uncertainty about the critical mass of $^{237}$Np in its oxide and metal forms, as seen in ANSI/ANS-8.15 (1981), "Nuclear Criticality Control of Special Actinide Elements." [3]

Knowing the precise critical mass of this element not only will help to validate storage limits or optimize storage configurations for safe disposition of these materials, but will also save thousands of dollars in transportation and disposition costs.

## 2. Description of the Experiment

### 2.1 Materials

The experiment was performed on the Planet vertical assembly machine at Los Alamos National Laboratory. The critical mass experiment consisted of surrounding the 6-kg $^{237}$Np sphere with highly enriched uranium (HEU) nesting shells until criticality was achieved. The neptunium sphere was 8.29 cm in diameter and its calculated density was 20.29 g/cm$^3$, which is very close to its theoretical density of 20.45 g/cm$^3$. The neptunium sphere weighed 6070.4 g and the chemical analysis of the sprue extension (see Fig. 1) showed that the sphere was 98.8 wt % neptunium, 0.035 wt % uranium, and 0.0355 wt % plutonium. There were also traces of americium found in the neptunium sphere. Table 1 shows the elements found in the chemical analysis of the sprue.

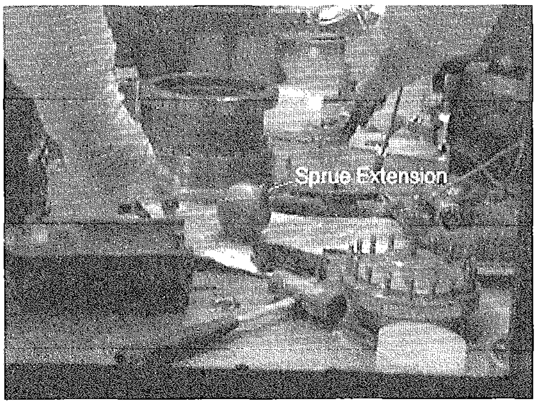

**Fig. 1** Bare neptunium sphere with the sprue.

To reduce the radiation exposure to the experimenters, the neptunium sphere was clad with a 0.261-cm thick layer of tungsten, and two 0.191-cm thick layers of nickel. The gamma radiation exposure at contact was 300 mR/h. The total weight of the sphere, including the clad materials, was 8026.9 g.

**Table 1.** Chemical analysis of neptunium sphere

| Element | wt % |
|---|---|
| $^{237}$Np | 98.8 |
| Uranium | 0.035 |
| $^{233}$U | 9.92 |
| $^{234}$U | 1.61 |
| $^{235}$U | 79.2 |
| $^{236}$U | 0.44 |
| $^{238}$U | 8.74 |
| Plutonium | 0.0355 |
| $^{238}$Pu | 4.45 |
| $^{239}$Pu | 88.18 |
| $^{240}$Pu | 6.32 |
| $^{241}$Pu | 0.17 |
| $^{242}$Pu | 0.89 |
| Element | ppm |
| Americium | |
| $^{241}$Am | 6.0 |
| $^{243}$Am | 1823.0 |

### 2.2 Experiment

The starting configuration consisted of the neptunium sphere and one or two HEU shells surrounding the neptunium sphere (see Fig. 2).

**Fig. 2** Neptunium sphere surrounded with HEU shells.

An aluminum spacer was used to perfectly accommodate the neptunium sphere in the first set of HEU shells. The HEU shells vary in accumulative weight from 1–66 kg. Four $BF_3$ detectors were used to monitor the neutron population from the assembly. A 1/M approach to critical was performed following the guidelines of existing operating procedures for the hand-stacking and remote operations. Once the hand-stacking limit was reached, the core shown in Fig. 3 was reconfigured so that approximately 33 105.1 g of HEU hemishells and the neptunium sphere were placed on the movable platen of the Planet vertical assembly machine. HEU hemishells were added to the top of the platform (see Fig. 4) and the Planet assembly was operated remotely. Experimenters continued adding hemishells of HEU to the top until criticality was achieved on September 24, 2002.

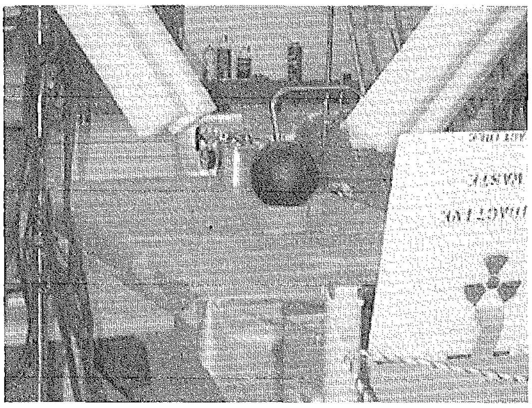

**Fig. 3** Hand-stacking operation of the $^{237}$Np sphere and HEU shells.

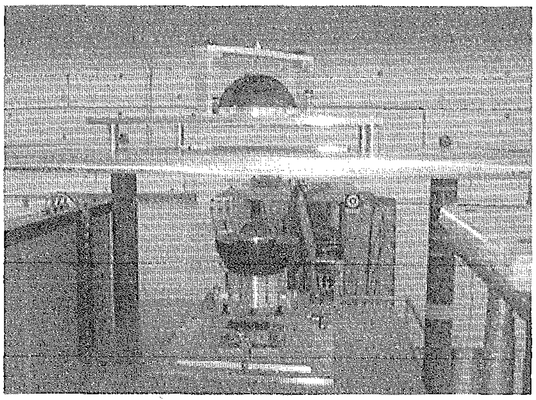

**Fig. 4** Final configuration of the $^{237}$Np/HEU experiment.

### 3. Results

A 1/M was plotted based upon the normalized counting rates as a function of uranium mass (see Fig. 5) and as a function of separation distance. A 0.32-cm-thick, 10-cm in diameter aluminum spacer was used between the top and bottom part of the core to limit the excess reactivity of the system. The total uranium mass was 62 555 g. When the two halves were fully closed with the aluminum spacer between them, a reactor period of 5.729 s was observed. This is equivalent to a $k_{eff}$ of 1.0026, assuming a calculated

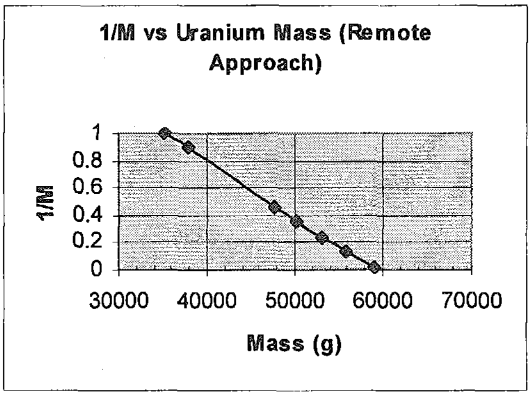

**Fig. 5** 1/M as a function of uranium mass.

$\beta_{eff}$ of 0.0054. An MCNP calculation of the same experiment yielded a $k_{eff}$ of 0.99089 ± 0.0003. It is believed that the discrepancy between the experiment and computer code could be explained by the inaccurate models of the inelastic scattering cross section of $^{237}$Np found in ENDF/B-VI. It was also noted by the Nuclear Physics group, T-16, at Los Alamos National Laboratory that the fission cross section, and ν (the average number of neutrons per fission) for $^{237}$Np need some improvement in the existing models.

## 4. Conclusion

Based on these preliminary results, it is estimated that the critical mass of $^{237}$Np for a bare system ranges from kilogram weights in the high fifties to low sixties. Finally, future experiments with various reflectors will tend to narrow a little bit more the uncertainty on the critical mass.

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