

# Measuring (n,f) cross-sections of short-lived states

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## Abstract

This paper reviews measurements of fission cross-sections of short-lived nuclear states, summarizes the formidable experimental difficulties involved and suggests novel methods of overcoming some of those difficulties. It is specifically concerned with the two such states that have been well characterized, the  $J^\pi = \frac{1}{2}^+$  (26 m) isomeric  $^{235m}\text{U}$  and the  $J^\pi = 1^-$  (16 h) ground state (shorter lived than the isomer)  $^{242gs}\text{Am}$ , and with measuring their fission cross-sections at MeV energies. These measurements are formidably difficult, partly because of the need to produce, separate and collect the short-lived states before they decay, and partly because of their comparatively small fission cross-sections at these energies. I present quantitative calculations of the efficiency of advection of recoiling  $^{235m}\text{U}$  isomers by flowing gas in competition with diffusive loss to the surface containing the mother  $^{239}\text{Pu}$ . This paper reports the initial development and evaluation of some of the methods that must be developed to make the experiments feasible.

# 1 Introduction

In order to test theories [1, 2, 3, 4] of fission cross-sections, it is desirable to measure the (n,f) cross-sections of both an isomeric state and the ground state of the same isotope of the same element. This would test the ability to calculate effects that depend only on spin and parity without the confusion of effects that depend on (Z,A). At present there is no isotope for which the (n,f) cross-section has been measured successfully at MeV energies, where theoretical predictions can be made. Measurements [5, 6, 7] exist only at thermal and cold energies, where the cross-section is expected to be dominated by incalculable resonances, so that it may not be a useful test of theoretical predictions.

The purpose of this paper is to estimate the parameters of the sources of the short-lived states and of their irradiation that would be required to obtain useful data. This is necessary to justify further investigation. Because these experiments would be difficult and ambitious, and (unlike the easier experiments with thermal neutrons) have never been attempted, bringing these ideas to the attention of the experimental nuclear physics and reactor communities will expose them to useful criticism and further their development to the point at which detailed experimental design would become appropriate.

There are two fissionable isotopes that have more than one level with a lifetime long enough to offer some hope of performing the measurement:

- $^{235}\text{U}$  has a 77 eV  $\frac{1}{2}^+$  excited state of 26 m half-life in addition to its long-lived (700,000,000 y)  $\frac{7}{2}^-$  ground state.
- $^{242}\text{Am}$  has a long-lived (141 y)  $5^-$  isomer and a short-lived (16 h)  $1^-$  ground state; this isotope has the unusual (but not unique) property that almost all of it is found in the isomeric state.

In each case the chief experimental obstacle is the brief lifetime of the shorter-lived state, making it difficult to accumulate a quantity sufficient for experimental measurement. In addition, direct separation of isomers from ground states has never been demonstrated (isomeric shifts of the hyperfine structure of atomic levels make it possible, in principle, by the methods of atomic vapor laser isotope separation) so that the shorter lived states must be separated at birth. Fortunately, separation at birth is possible.

The experimental and technical difficulties are formidable. Most of them have been considered before, but the earlier literature, which I cite whenever possible, is incomplete because unpromising or unsuccessful work is often not documented. This paper includes a review of an “underground” of work on these problems. It also proposes and calculates as well as is feasible with present understanding of the difficulties. Yet, with sufficient resources and determination, it may be possible to overcome these obstacles.

This paper is the full report of the work presented [8] at the November, 2009 meeting of the American Nuclear Society in Washington, D. C.

## 2 $^{235m}\text{U}$

$^{235m}\text{U}$  is the product of the 5.245 MeV alpha-decay of  $^{239}\text{Pu}$ , with a recoil energy of 89 KeV. More than 99.9% of the decays are to the isomer. Hence  $^{239}\text{Pu}$  is a steady source of the uranium isomer. However, because of its rapid decay the isomer soon becomes a trace constituent of the ground state  $^{235}\text{U}$ . In order to do experiments on the isomer, it must be separated from the plutonium source in a few tens of minutes or less.

One possible approach would be to chemically purify the plutonium, removing the uranium that has built up, and then repurify, extracting the newly created isomer perhaps 10 minutes later. This is a formidable problem in radiochemistry but has been solved on a small scale [6]. The first purification must completely extract all the uranium, which will have been in the ground state (the tolerable fraction of the uranium remaining is less than the ratio of the isomeric half life to the time since fabrication of the sample, or  $< 0.0006$  for a month-old sample), and the second purification, taking no longer than about 10 minutes, must extract 2 ppb of newly produced (isomeric) uranium from plutonium. We estimate in Section 5 that to measure the cross-section at MeV energies will require isolation of about 100 times as many isomeric nuclei as achieved by [6] for their measurements at thermal energies.

Another approach uses the recoil of the newly formed  $^{235m}\text{U}$  to separate it from the plutonium. This recoil permits the  $^{235m}\text{U}$  to penetrate a small,

and uncertain, thickness of parent metallic Pu or Pu compound. Measurements [9] on other  $\alpha$ -decay recoil nuclei suggest a stopping column density  $\approx 20\mu\text{g}/\text{cm}^2$ , and therefore a stopping length  $\ell \approx 0.01\mu$  in metallic plutonium. The  $^{235m}\text{U}$  escaping from the plutonium may be collected electrostatically [5, 7] or swept up in a stream of gas [10], possibly containing aerosols to which it would be attracted and bound if ionized [7], and collected.

Previous attempts to collect  $^{235m}\text{U}$  from gas flowing through a tube coated with Pu have not succeeded [6, 11], perhaps because diffusion in the carrier gas permitted the  $^{235m}\text{U}$  to deposit on unintended surfaces, or because flow of the carrier gas swept it around the intended collector. Free atoms will stick to almost any surface they encounter. The first problem is acute at low gas densities (where the diffusivity is large) and the second is acute at high gas densities (where the diffusivity is small).

For the mother isotope of number density  $n_m$  and half-life  $t_m = 24,100\text{ y}$ , and the daughter with half-life  $t_d = 26\text{ m}$ , collecting the daughter for a time  $t$  produces a yield

$$Y_d = \frac{1}{2}n_m\ell\frac{t_d}{t_m}(1 - 2^{(-t/t_d)}) = 1.5 \times 10^7 \frac{\ell}{10^{-6}\text{ cm}} \text{ cm}^{-2}, \quad (1)$$

where the leading factor of  $1/2$  is the fraction of upward directed daughters and we have taken  $t = t_d$  and  $n_m = 4 \times 10^{22}\text{ cm}^{-3}$  for  $\delta$ -plutonium. The parenthesis then introduces another factor of  $1/2$ ; longer collection increases this factor slowly, but at the price of growing, eventually dominant,

contamination with the ground state.

To produce  $N = 4 \times 10^{12}$   $^{235m}\text{U}$  nuclei, as estimated in Section 5 for the irradiation target, would require a plutonium source of area  $30 \times (10^{-8} \text{ m}/\ell)$   $\text{m}^2$ . This may be feasible, but is challenging; the source could consist of long porous tubes through whose walls the carrier gas would flow.

In any method of producing  $^{235m}\text{U}$  speed is of the essence because of its 26 minute half-life. Talbert, *et al.* [6] have shown that it is possible to perform complex chemical extractions and purifications, followed by sample preparation and irradiation, within these time limits. Even when recoil is used to separate the  $^{235m}\text{U}$  from its mother  $^{239}\text{Pu}$ , the isomer will be accompanied by some sputtered plutonium. This can be removed chemically along with the separation of the isomer from the material on which it is collected.

## 2.1 Flow-through sources and collectors

Rather than trying to find an optimum carrier gas density, we propose an alternative solution, flowing gas through a porous source and a porous collector. The principle is that of a flyswatter: A porous object admits the passage of gas, while trapping particles suspended in it. The microphysics is different, relying on diffusion of atomic species to solid surfaces (the atomic species are smaller than the pores, unlike the fly). For example, deposit the plutonium source on the downstream surface of a nuclepore (polycarbonate) filter through which carrier gas will flow, being careful that the pores remain unblocked. The configuration is illustrated in Figure 1.

To determine feasibility, we first estimate the stopping length of  $^{235m}\text{U}$  in the *gas*. It loses about half its momentum each time it passes through a column density of gas with mass equal to its own mass divided by a collision cross-section. Hence its stopping (thermalization) length

$$\ell_s \approx \frac{\ln(E_{recoil}/k_B T)}{2 \ln 2} \frac{m_U}{\rho_{gas} \sigma_{coll}} \approx 10 \frac{m_U}{\rho_{gas} \sigma_{coll}}, \quad (2)$$

where  $\rho_{gas}$  is the gas density,  $\sigma_{coll}$  the effective collision cross-section, which we take to be  $10^{-19} \text{ m}^2$  ( $10^{-15} \text{ cm}^2$ ), and room temperature gas is assumed. The angle-averaged injection distance is  $\ell_s/2$ . This is generally quite short;  $\rho_{gas} \ell_s/2 \approx 2 \times 10^{-6} \text{ g/cm}^2$ .

Collection is achieved by passing the gas through another filter or a porous sacrificial medium. The  $^{235m}\text{U}$  will deposit on its front surface and the inside of its pores, and may be separated by chemical processing of the entire collector. If the pores are narrow the collector need not be thick because of the rapid diffusion of the  $^{235m}\text{U}$  atoms through the gas to the walls of the pores. If they attach to aerosols in the gas flow, the aerosols may be trapped by the pores.

## 2.2 Collection efficiency

We calculate the collection efficiency of  $^{235m}\text{U}$  injected into a carrier gas flow from a porous surface through which the gas penetrates. The results are very generally applicable to any problem in which there is competition between



advection of some species from a volume source (in this case, the volume is the region through which isomers recoiling from the solid plutonium-containing surface are stopped in the gas) and diffusion of that species to a surface to which they stick.

The fundamental equation is that of conservation of particles:

$$\frac{\partial n}{\partial t} + V \frac{\partial n}{\partial x} - D \frac{\partial^2 n}{\partial x^2} - S(x) = 0, \quad (3)$$

where  $n(x, t)$  is the density of the particles, we assume planar symmetry and steady uniform gas flow at the speed  $V$  in the  $+x$  direction,  $D$  is the diffusion coefficient of the particles in the gas and  $S(x)$  is their source strength. We consider only stationary solutions, and replace partial derivatives with respect to  $x$  by ordinary derivatives:

$$V \frac{dn}{dx} - D \frac{d^2n}{dx^2} - S(x) = 0. \quad (4)$$

For an isotropic source of particles with stopping length  $\ell_s$  the volume source is readily seen to be uniformly distributed in  $x$ :

$$S(x) = \begin{cases} S_0/(2\ell_s) & x \leq \ell_s \\ 0 & x > \ell_s, \end{cases} \quad (5)$$

where  $S_0$  is the areal source strength (particles per unit area per unit time) and the factor of 2 comes from the fact that half the particles are emitted in

the  $-x$  direction and do not enter the carrier gas.

Writing  $y \equiv dn/dx$  for convenience, the only physically meaningful solution for  $x > \ell_s$  where  $S(x) = 0$  is  $y = 0$ ; any other solution would diverge exponentially as  $x \rightarrow \infty$ . Then for  $x > \ell_s$  the density of isomers  $n(x)$  is constant:

$$n(x) = n_\infty, \quad (6)$$

where the flux (per unit area) is  $n_\infty V$ .

For  $0 \leq x \leq \ell_s$  Eq. 4 becomes,

$$\frac{dy}{dx} = \frac{V}{D}y - \frac{S(x)}{D}. \quad (7)$$

This is readily integrated:

$$y = C \exp(Vx/D) - \frac{S_0}{2\ell_s V} \exp(Vx/D) (1 - \exp(-Vx/D)). \quad (8)$$

The constant of integration  $C$  is determined by requiring continuity of  $y$  at  $x = \ell_s$  where  $y = 0$ :

$$C = -\frac{S_0}{2\ell_s V} (\exp(-\alpha_\ell) - 1), \quad (9)$$

where we have defined the parameter  $\alpha_\ell \equiv V\ell_s/D$ . Then for  $0 \leq x \leq \ell_s$

$$y = -\frac{S_0}{2\ell_s V} (\exp(V(x - \ell_s)/D) - 1). \quad (10)$$

Eq. 10 is readily integrated to give  $n(x)$  for  $0 \leq x \leq \ell_s$ :

$$n(x) = -\frac{S_0 D}{2\ell_s V^2} \exp(-\alpha_\ell) (\exp(Vx/D) - 1) + \frac{S_0 x}{2\ell_s V}, \quad (11)$$

where we have used the boundary condition, appropriate to a surface that is a perfect sink (any isomers diffusing to it stick to it, as will generally be true of free atoms or ions)  $n(0) = 0$ . Defining  $\alpha \equiv Vx/D$ , and  $n_0 \equiv S_0/2V$  (the isomer density in the carrier gas if there were no losses by diffusion to the surface), Eq. 11 can be rewritten

$$n(x) = \frac{n_0}{\alpha_\ell} [\alpha - \exp(-\alpha_\ell) (\exp(\alpha) - 1)]. \quad (12)$$

For  $x > \ell_s$   $n(x) = n_\infty$ , where  $n_\infty$  may be found by continuity, taking  $x = \ell_s$  ( $\alpha = \alpha_\ell$ ) in Eq. 12:

$$n_\infty = n_0 \left( 1 - \frac{1 - \exp(-\alpha_\ell)}{\alpha_\ell} \right). \quad (13)$$

This has the limits

$$\frac{n_\infty}{n_0} = \begin{cases} \frac{\alpha_\ell}{2} - \frac{\alpha_\ell^2}{6} + \dots & \alpha_\ell \rightarrow 0 \\ 1 - \frac{1}{\alpha_\ell} + \dots & \alpha_\ell \rightarrow \infty. \end{cases} \quad (14)$$

The fraction of the recoiling  $^{235m}\text{U}$  that are swept into the gas flow is the collection efficiency  $\epsilon \equiv n_\infty/n_0$ . The remaining isomers diffuse back to

the source and stick to its surface. Using our previous estimate 2 for  $\ell_s$  and approximating  $D \approx v_{thU} m_U / (3\rho_{gas} \sigma_{coll})$ , where  $v_{thU}$  is the thermal velocity of a uranium atom, we obtain

$$\alpha_\ell \approx 30 \frac{V}{v_{thU}}. \quad (15)$$

These results are summarized in Fig. 2. For example, the condition  $\alpha_\ell > 2$  ( $\epsilon > 0.57$ ) corresponds, at room temperature, to  $V > 7$  m/s. This is not difficult to provide, and implies a very low Mach number and little dissipation in the gas flow.

### 3 $^{242gs}\text{Am}$

$^{242gs}\text{Am}$  is produced by internal transition of  $^{242m}\text{Am}$ . At least one electron (usually from the L shell) is ejected, ionized by the 49 KeV of the excited state, and more may be ejected as the hole is filled by a cascade. Hence the ground state product is ionized and may be collected electrostatically, just as for  $^{235m}\text{U}$  [5, 7], as shown in Figure 3. Subsequent isolation of the short-lived ground state requires only chemical separation from the collecting electrode and from its (chemically distinct) decay products.

Suppose we wish to collect  $N$   $^{242gs}\text{Am}$  nuclei from a region of area  $A$  between two capacitor plates, separated by a distance  $d$ , in a time  $t = t_{gs} 1/2 / \ln 2$ , after which 63% of the asymptotic limit has been collected (longer

collections contaminate the collecting cathode with fissionable  $^{242}\text{Pu}$  and  $^{242}\text{Cm}$ , although these can be removed chemically when the  $^{242\text{gs}}\text{Am}$  is extracted, unlike the otherwise analogous contamination of  $^{235\text{m}}\text{U}$  by  $^{235\text{gs}}\text{U}$ ). The volume  $Ad$  is filled with  $^{242\text{m}}\text{Am}$ , as some suitable vapor at number density  $n_v$ , and decays at a rate  $\nu_m$ . The density and separation are limited by charge exchange, with cross-section  $\sigma_{ce}$ , between the ionized  $^{242\text{gs}}\text{Am}$ -containing species and the neutral  $^{242\text{m}}\text{Am}$  species, so that  $n_v d \lesssim 1/\sigma_{ce}$ , with a strong inequality desirable. The ground state production rate must be at least  $N/t$ , from which we obtain the minimum area

$$A > \frac{N}{t\nu_m n_v d} > \frac{N\sigma_{ce}}{t\nu_m} = N\sigma_{ce} \frac{t_{m1/2}}{t_{gs1/2}} \approx 80 \frac{N}{10^{11}} \text{ cm}^2, \quad (16)$$

where we have taken  $\sigma_{ce} = \mathcal{O}(10^{-18} \text{ m}^2)$  ( $\mathcal{O}(10^{-14} \text{ cm}^2)$ ), generally a conservative upper bound to atomic and molecular resonant  $\sigma_{ce}$ . The required  $A$  is feasible if  $N \lesssim 10^{12}$ , as estimated in Section 5.

The assumed  $n_v d < 10^{18}/\text{m}^2$  ( $10^{14}/\text{cm}^2$ ) suggests densities  $n_v = 10^{19}$ – $10^{20}/\text{m}^3$  ( $10^{13}$ – $10^{14}/\text{cm}^3$ ) in order to keep the electrode separation in the convenient range 1–10 cm. At temperatures  $\mathcal{O}(10^3)$  °K this range corresponds to pressures  $\mathcal{O}(0.1\text{--}1 \text{ Pa})$  ( $1\text{--}10 \text{ dyne/cm}^2$ ,  $10^{-3}\text{--}10^{-2} \text{ mm-Hg}$ ).

Americium fluorides are not volatile (unlike  $\text{UF}_6$ ). The most volatile fluoride,  $\text{AmF}_4$  [12], disproportionates ( $\text{AmF}_4 \rightarrow \text{AmF}_3 + \frac{1}{2} \text{F}_2$ ) above about 900 °K, at which  $n_v \approx 2 \times 10^{18}/\text{m}^3$  ( $p \approx 10^{-4} \text{ mm-Hg}$ ) [13].  $\text{AmF}_4$  vapor is very corrosive because of its readiness to fluorinate even non-reactive metals.

AmF<sub>3</sub> is significantly less volatile than AmF<sub>4</sub>. A vapor pressure of 10<sup>-2</sup> mm-Hg requires  $T \approx 1500^\circ\text{K}$  [14, 15], while even 10<sup>-4</sup> mm-Hg, obtainable with AmF<sub>4</sub>, requires  $T \approx 1300^\circ\text{K}$ . For metallic americium the corresponding temperatures are about 1260°K and 1060°K, respectively, lower than those required for the trifluoride [16]. In addition, it has no “chemistry” to deal with.

It might be possible to use tetrafluoride vapor, but a larger gas volume  $Ad$  would be required than for higher gas densities. If  $N$  is as small as  $\mathcal{O}(10^{11})$ , as suggested in Section 5, than the lower vapor densities (than the upper limits permitted by charge exchange) permitted by the AmF<sub>4</sub> disproportionation temperature limit may be acceptable, while keeping the total source volume within bounds. Then

$$Ad \gtrsim \frac{N t_m}{n_v t_{gs}} \approx \frac{N}{10^{11}} \frac{10^{18} \text{ m}^{-3}}{n_v} 8 \times 10^{-3} \text{ m}^3. \quad (17)$$

This appears feasible, but depends on the chemical stability of AmF<sub>4</sub> vapor over times of several hours at 900°K. The vapor could be replenished continually, and later reconstituted from its disproportionation products, but this would add an additional complication, and raise the danger of loss of scarce material.

Americium organometallic compounds and oxides are much more promising because of their greater volatility [17, 18, 19, 20]. Quantitative measurements do not appear to exist, but temperatures  $\approx 130^\circ\text{C}$  may provide

sufficient vapor density.

The source dimensions scale according to the two constraints:

1. Loss of  $^{242\text{gs}}\text{Am}$  sets an upper bound  $d \lesssim 10^{14} \text{ cm}^{-2}/n_v$ .
2. Providing sufficient  $^{242\text{gs}}\text{Am}$  sets the lower bound (17) on the source volume. Higher vapor density  $n_v$  (requiring higher temperature) would permit smaller vapor volumes, but would not reduce the minimum collecting area (16).

These constraints are essentially independent of the vapor species, but the required temperature depends, of course, on the equilibrium vapor pressure.

## 4 The Neutron Source

Measurement of the (n,f) cross-section at MeV energies requires an intense source of MeV neutrons. This may be obtained by conversion of the thermal neutron flux of a reactor. Incident upon a shell of 19.9% enriched  $^{235}\text{U}$  of thickness  $\delta$ , as shown in Figure 4, a fraction (independent of the shell radius as long as it is  $\gg \delta$ )

$$f_{fiss} = 1 - E_2(n_{235}\delta\sigma_{th(n,f)235}) \quad (18)$$

of the thermal flux produces fissions in 19.9% enriched uranium, where  $n_{235} = 9.6 \times 10^{27} \text{ m}^{-3}$  ( $9.6 \times 10^{21} \text{ cm}^{-3}$ ),  $\sigma_{th(n,f)235} = 580 \text{ b}$  is its thermal neutron fission cross-section and  $E_2$  is the exponential integral. For  $\delta = 0.2 \text{ cm}$  the

argument of  $E_2$  is 1.12,  $f_{fiss} = 0.87$  (most of the thermal neutrons are incident obliquely and have much longer paths in the shell than at normal incidence) and the fission neutron flux is  $\nu f_{fiss} \approx 2.0$  times the incident thermal neutron flux where  $\nu$  is the thermally induced fission neutron multiplicity.

The attenuation of the fission neutrons in the uranium shell is readily estimated. The scattering probability (the cross-section is about 7 b on both uranium isotopes) is 0.07 on a radial path. Integrating over the isotropic source spectrum yields a transmission probability of 0.78. For the illustrated geometry we are interested only in nearly radial fission neutrons (because the target to be irradiated is near the center of the sphere), and their transmission probability is 0.93.

The residual thermal neutron flux must be absorbed so that it induces no more than a few percent as many fissions as the fast fission neutrons. This is readily accomplished by filling the inside of the enriched uranium shell with boron or boron carbide, possibly enriched with  $^{10}\text{B}$ . A thickness of 8 mm of natural (20%  $^{10}\text{B}$ )  $\text{B}_4\text{C}$ , pressed to solid density ( $\rho = 2.52 \times 10^3 \text{ kg/m}^3$ , 2.52 gm/cm<sup>3</sup>), provides 54  $e$ -folds of attenuation.

Ten  $e$ -folds are sufficient to reduce the thermal flux to levels at which it induces, for plausible assumptions as to the cross-sections, only a few percent as many fissions in the targets as the fission-spectrum neutrons of interest.  $^{10}\text{B}$ , unlike the resonance absorbers Cd and Gd, is also effective in attenuating the epithermal neutrons present in reactors.

Attenuation of the desired fission (MeV) neutrons by the  $\text{B}_4\text{C}$  is readily



estimated. Their total cross-section with either boron isotope or carbon is about 2 b, and the fraction surviving the 8 mm radial path is about 80%.

The achievable neutron flux is limited, not by the capabilities of research reactors, but by the heat load of the fissions in the enriched uranium shell. A thermal neutron flux of  $10^{16}/(\text{m}^2 \text{s})$  ( $10^{12}/(\text{cm}^2 \text{s})$ ) produces a fission power of  $250,000 \text{ W}/\text{m}^2$  ( $25 \text{ W}/\text{cm}^2$ ). This is a few times the heat flux of a kitchen stove on its high setting, and can be dissipated by free convection if immersed in water, or by forced gas flow, but represents a rough upper limit to the tolerable flux.

The thermal neutron calculations in this section are necessarily approximate because they do not allow for the evolution of the neutron spectrum as it is attenuated by processes ( $^{10}\text{Be}(n,\alpha)^7\text{Li}$  and  $^{235}\text{U}(n,\text{fiss})$ ) whose cross-sections are energy dependent (generally  $\propto E^{-1/2}$  at thermal energies). More accurate results, as well as inclusion of the epithermal neutrons present in reactors, require the straightforward use of a Monte Carlo code such as MCNP.

## 5 Required Target Quantities

If  $N_t$  target nuclei with fission cross-section  $\sigma_{n,\text{fiss}}$ , that decay with half-life  $t_{1/2}$ , are irradiated by a neutron flux  $F$  for a time  $t$ , the number of fissions will be

$$N_{\text{fiss}} = N_t F \sigma_{n,\text{fiss}} (1 - \exp(-t \ln 2 / t_{1/2})). \quad (19)$$

Although it might seem advantageous to take  $t \gtrsim t_{1/2}$ , that would confound the fissions of the intended targets with those of their decay products ( $^{235}\text{U}$  for  $^{235m}\text{U}$  and  $^{242}\text{Pu}$  and  $^{242}\text{Cm}$  for  $^{242gs}\text{Am}$ ) that are themselves fissionable. The decay products have cross-sections comparable to those of the intended targets, but that are imperfectly known so that accurate subtraction would not be possible. Hence it is necessary to take  $t \ll t_{1/2}$ , and here we adopt  $t = 0.1t_{1/2}/\ln 2$ .

In order to limit the contribution of counting statistics to the error in the measured cross-section to 3%,  $N_f = 1000$  would be required, noting that each fission produces two oppositely directed tracks; for a target plated on nuclear emulsion the counting efficiency may approach unity. Then

$$N_t = 7 \times 10^{15} \frac{N_f}{1000} \frac{\sigma_{n,fiss}}{1 \text{ b}} \frac{10^{12} \text{ cm}^{-2} \text{ s}^{-1}}{F} \frac{1 \text{ s}}{t_{1/2}}. \quad (20)$$

We use 1 b as a nominal cross-section for estimating the required target mass. Predicted (theoretical) isomeric fission cross-sections [1, 2, 3, 4] are typically about this value for MeV neutrons, so it sets a realistic scale for the required target masses. Substituting the appropriate half lives and taking the fractions to be unity yields

$$N_t = \begin{cases} 4 \times 10^{12} & ^{235m}\text{U} \\ 1.2 \times 10^{11} & ^{242gs}\text{Am}. \end{cases} \quad (21)$$

If the targets are plated as a monolayer, the areas required are each

less than  $1 \text{ mm}^2$ , and several targets can be placed near the center of the neutron source, as illustrated. Of course, thicker targets can be used, with a corresponding reduction in area, so long as they are thin compared to the fission product stopping length in high-Z material of several  $\mu$ .

The quantity of source material required is

$$N_{source} = N_t \frac{t_{1/2 source}}{t_{1/2}} = \begin{cases} 2 \times 10^{21} \text{ (0.8 g)} & {}^{239}\text{Pu} \\ 9 \times 10^{15} \text{ (4 } \mu\text{g)} & {}^{242m}\text{Am.} \end{cases} \quad (22)$$

Although the availability of  ${}^{242m}\text{Am}$  is limited, the quantity required is very small;  ${}^{239}\text{Pu}$  is, of course, available in essentially unlimited quantity.

## 6 Counting the Fissions

If the targets are plated upon nuclear emulsions, fissions occurring during irradiation may be measured at leisure by counting etched tracks. This has the advantage that counting is done in a low-radiation environment, with an efficiency approaching 100%, although it is painstaking work through a microscope. D'Eer, *et al.* [7] counted fissions during irradiation by coincident detection of fragments by two semiconductor detectors, with the disadvantage of working in a very high radiation environment. In either case, a statistically 3% accurate cross-section would require counting only 1000 fissions.

Fissions are high energy events that produce a characteristic signature. Their energetic but low speed products deposit energy at a uniquely high

rate per unit length of their path; there is essentially no background. In a nuclear emulsion their tracks are unmistakable. A particular advantage is that multiple targets, both signal and calibration, can be irradiated simultaneously and in close proximity (less than a mm apart) to ensure that they are exposed to the same neutron flux, but with no ambiguity as to which track is associated with which target because the tracks enter the emulsion immediately under the targets.

## 7 Calibration

The proposed experiments would need calibration of the thermal neutron flux at the irradiated targets. This could be provided by a control sample of  $^{235}\text{U}$ , and would amount to normalizing the measured cross-sections to the (well known) thermal neutron fission cross-section of  $^{235}\text{U}$ . The flux of MeV neutrons would be monitored (it is intended to be very small) by a control sample of  $^{238}\text{U}$  or  $^{232}\text{Th}$  with well known fission cross-sections at that energy but essentially zero cross-section at thermal energy. In the test samples differential measurements (using different periods of decay of the short-lived states) would be required to allow for the effects of isotopic or isomeric impurity.

The quantities of the irradiated targets would be determined after their natural decay. The mass of  $^{235m}\text{U}$  could be determined by measuring the fission rate of its daughter  $^{235}\text{U}$  in an independently calibrated thermal neu-

tron flux. The mass of  $^{242gs}\text{Am}$  could be determined by counting the 44 KeV (0.03%) gamma rays or the  $\alpha$  decays of its (82.7% branching ratio, 163 d) daughter  $^{242}\text{Cm}$  following chemical separation.

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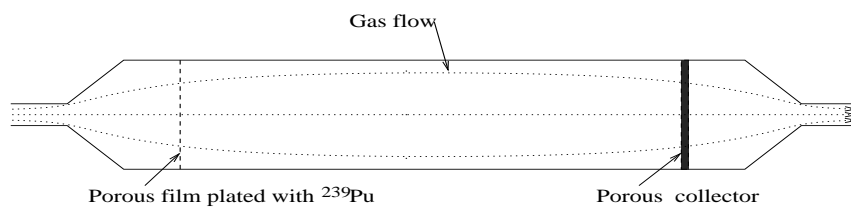
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## 8 Figure Captions

1. Schematic of  $^{235m}\text{U}$  source. Inert gas flows through the tube from left to right, passing through pores in the source and collector. The source has a thin layer of  $^{239}\text{Pu}$  plated on a supporting film, with gas flowing through it fast enough (see text) that  $^{235m}\text{U}$  that stop in the gas do not diffuse back to the film. The  $^{235m}\text{U}$  is swept by the gas flow to the porous collector where it diffuses to and attaches to the surface or to the walls of its pores, for later chemical extraction.
2. Upper graph shows the density of  $^{235m}\text{U}$  isomers as a function of distance from a porous source through which gas flows at a speed  $V$ . The distance is normalized to the recoil stopping length in the gas  $\ell_s$  and the density normalized to  $S_0/2V$ , where  $S_0$  is the source strength; this would be the density were there no diffusive losses of isomers to the surface of the source. Lower graph shows the efficiency of entrainment of isomers in the gas flow as a function of the parameter  $\alpha_\ell \equiv V\ell_s/D$ , where  $D$  is the diffusivity of isomers in the gas; this describes the comparative rates of advection of isomers from their source to that at which they diffuse back to it, the source surface acting as a sink.
3. Apparatus for electrostatic collection of  $^{242gs}\text{Am}$  from vapor. The vapor is indicated as  $\text{AmF}_4$ , but it could be atomic Am or  $\text{AmF}_3$ .
4. Thermal to fission spectrum neutron converter, approximately to scale.





$^{235}\text{mU}$  Source and Collector

Figure 1:

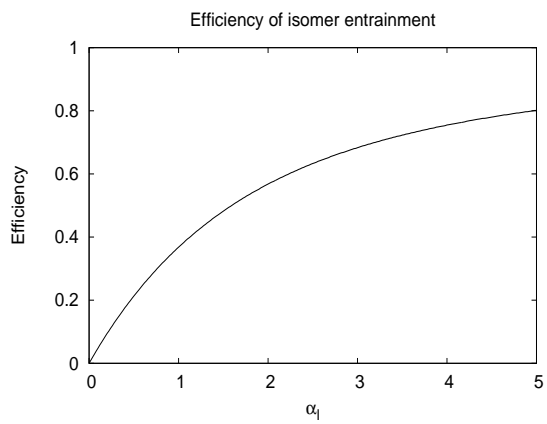
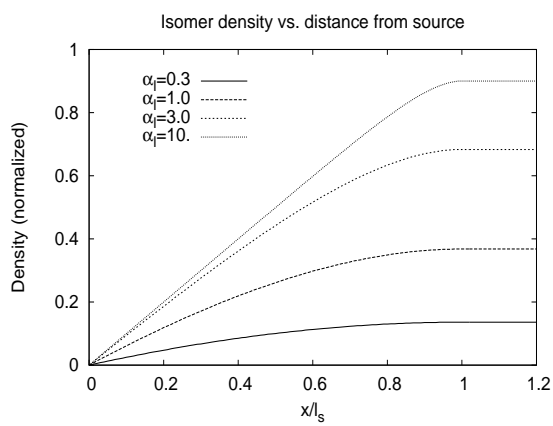
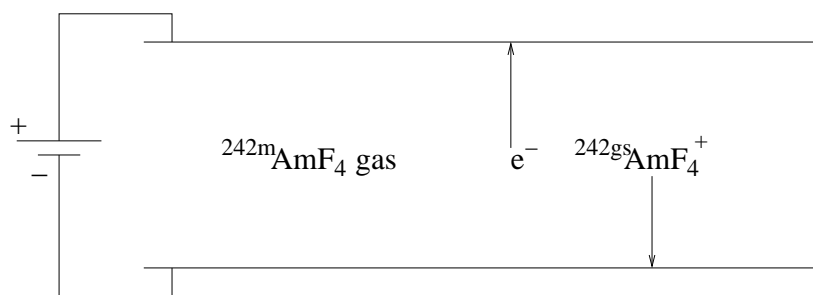


Figure 2:



Collection of ground state from isomer vapor

Figure 3:

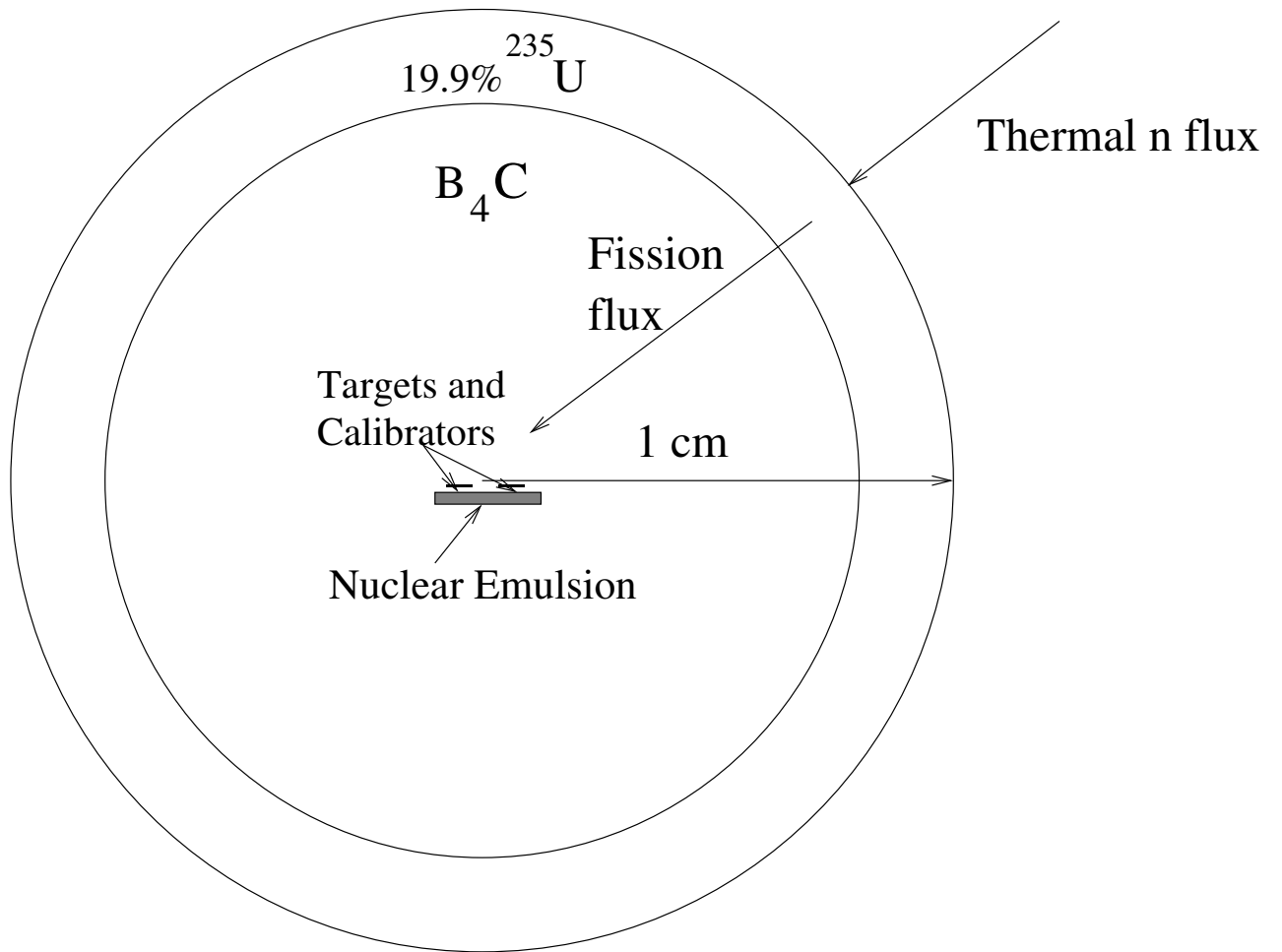


Figure 4: