Neutron diffusion geometry and criticality

Mathews and Walker, problem 8.2 [1]

Neutrons in a fissionable material obey the modified diffusion equation

$$\frac{\partial n}{\partial t} + \boldsymbol{\nabla}\cdot(-D_0 \boldsymbol{\nabla} n) = \chi n \tag{1}$$

where $n(\mathbf{r}, t)$ is the number density and D_0 is the diffusion constant. Were $\chi = 0$, using the divergence theorem this would be the statement of conservation of neutron number; finite positive χ indicates a *source* of neutrons within the material, proportional to what's already there. Knowledge that χ could become positive (the fission of a nucleus on average produced more neutrons than the one 'used up' to produce the splitting, together with 200 MeV of energy) gave rise to nuclear power and nuclear weapons.

For fissionable materials χ and D_0 may depend very strongly on the energy of the neutrons. For weapons use, only high-energy neutrons, released from fission, are relevant. For power reactors, 'thermal' neutrons, with energies of fractions of an eV, are relevant.

It is always important to bear in mind that the permissible mathematical 'chunks' of the solution to the equations above are determined strictly by the PDE, but the actual solution is found by imposing boundary and initial conditions. This fixes the coefficients in the mixture of solutions for our specific physical problem.

Spherical geometry

As is well known, the shape in three dimensions which encloses the largest volume for a given surface area is a sphere. This in turn implies this shape requires the least amount of (extraordinarily expensive) fissile material and mandates using spherical polar coordinates. As we will see, the boundary conditions on the neutron density will determine the specific combinations of solutions of the partial differential equation. Using separation of variables, the general solution is of the form

$$n(r, t) = R(ar)\, e^{\left(\chi - a^2 D_0\right)t}. \tag{2}$$

Notice that there is no net time dependence in the neutron number at *criticality*, when $a^2 = \chi / D_0$. If a is too small, the neutron number will grow exponentially in time.

As a first guess, we might assume that there are no neutrons on and beyond the surface of the sphere. For a sphere

$$R(ar) = j_0(ar) = \frac{\sin ar}{ar} \tag{3}$$

where j_0 is the zeroth-order spherical Bessel function (well-behaved for all radii). If we require than $n(r = R_0) = 0$ (for sphere radius R_0) then a must be π / R_0 for the first solution (the first radial occurrence of a zero). This determines $n(r,t)$ and tells us that the critical mass can be found from (at criticality)

$$R_0 = \pi/a = \pi\sqrt{\frac{D_0}{\chi}}. \tag{4}$$

As a second guess, we could assume that there is no *current* of neutrons radially outward through the sphere surface. Since

$$\hat{\mathbf{r}}\cdot\boldsymbol{\nabla} n(r,t) = \frac{\partial n}{\partial r}, \tag{5}$$

setting the current to zero becomes a (Neumann) boundary condition for the diffusion equation. We then find that the lowest value of aR_0 must be the first zero of the function $(x - \tan x)$, about 4.49341.

The radial dependence for the case of zero density and zero derivative on the sphere boundary is shown in in Fig. 1. While both are mathematically valid, only the zero density case is physically relevant, simply because solutions of almost all diffusion problems must be positive definite–there is no such thing as a negative density. Thus there is no solution if requiring the surface current to be zero.

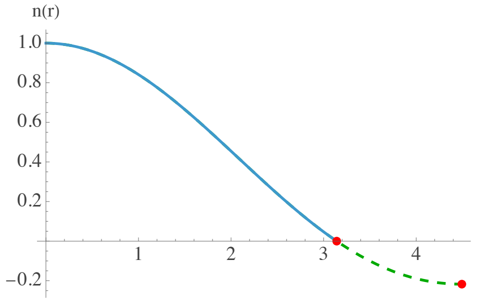

Figure 1: Radial dependence of neutron density if density is zero (blue curve) or derivative (dashed green) is zero on surface. Red dots indicate point at which boundary condition is satisfied, defining critical radius. Density must be positive, so the dashed green curve is mathematically but not physically valid.

Finally, we consider a boundary condition based on the application of 'kinetic theory' to neutrons near the surface of the sphere. It is

$$n\left(R_0\right) = -\frac{2}{3}\lambda_t\left(\frac{dn}{dr}\right)_{R_0}. \tag{6}$$

See the appendices of Reed [2] for a careful derivation.

The physics: there is a finite neutron density on the sphere surface if there is a radial current out through the surface. We expect such a current from neutrons escaping from the sphere; none are coming in. It is 'as if' this outward current has 'expanded' the radius of the sphere. This in turn permits the *actual* radius of the sphere to be smaller at criticality.

This is called a *Robin* boundary condition, arising also in convective heat flow.

We will use Eq. 6 as a *boundary condition*. If we expand near the surface ($r = R_0$)

$$dn(R_0) \simeq -\frac{3}{2\lambda_t} n(R_0)\, dr, \tag{7}$$

whose solution is

$$n(r \geq R_0) = n\left(R_0\right) e^{-\frac{3}{2}\frac{r-R_0}{\lambda_t}}. \tag{8}$$

As noted by John H. Bickel [3], the secret is to "use transport theory to make diffusion theory work in specific areas where it would be expected to fail." This 'secret' must have been known to Manhattan Project scientists even though the field of neutron transport was very new, since the neutron had been discovered only in 1932.

Thus the neutron density spills outside the sphere radius but decays exponentially with a characteristic length of 2/3 of the transport mean free path λ_t. Since (see table) both R_0 and λ_t are several centimeters, we expect this boundary condition to have a significant impact on criticality.

If we define $Q \equiv \frac{3}{2\lambda_t a}$ then the criticality condition with these 'effusion' boundary conditions is given by the first zero Z_{efs} of the function

$$x \cot x + Qx - 1 \tag{9}$$

where $x = ar$. We find the critical radius R_0 at this Z_{efs} via $R_0 = Z_{efs}/a$. Thus the price we pay for a reliable boundary condition appears to be the introduction of a new parameter, the 'transport [or total] mean free path' λ_t. But in fact, all parameters in the diffusion equation, a **macroscopic** theory, are already mixtures of statistical averages of **microscopic** properties. So far the diffusion equation has been strictly phenomenological, with unknown parameters χ and D_0. In fact, diffusion itself is a macroscopic phenomenon which depends on describing particle density as a continuum, instead of via a microscopic distribution of discrete particles, such as

$$n(\mathbf{r}, t) = \sum_{i=1}^{N} \delta\left(\mathbf{r} - \mathbf{r}_i(t)\right) \tag{10}$$

where the ith of N particles is found at position $\mathbf{r}_i$ at time t. A continuous description via $n(\mathbf{r}, t)$ requires that we consider length scales much larger than the mean distance between particles. *Kinetic theory* was developed in the mid 19th through early 20th centuries. 'Mean free paths'–the statistically averaged distance a neutron travels before experiencing an event–can be defined for several processes. Important parameters include

$$\begin{aligned} D_0 &= \frac{1}{3}\lambda_t \langle v \rangle & (11) \\ \chi &= \frac{\langle v \rangle}{\lambda_f}(\nu - 1), & (12) \end{aligned}$$

Here ν is the number of neutrons produced per average fission event, $\langle v \rangle$ is the average speed of a neutron, λ_t is the total mean free path, and λ_f is the mean free path for a neutron to induce a fission event. We could also introduce *time* scales from length scales, such as the mean time a neutron must travel before causing a fission, via $\tau = \lambda_f/\langle v \rangle$, etc. Any quantity in the table below which involves seconds is quite large, essentially due to the high average neutron speeds. A 'thermal neutron' has a kinetic energy of about $\frac{3}{2}k_B T \simeq \frac{1}{40}$ eV. By contrast, neutrons produced from a fission reaction have kinetic energies 1-20 MeV or so (speeds 1.4-4.8$\times 10^9$ cm/sec). With characteristic length scales of cm, time scales are nanoseconds; thus D_0 and χ are very large. Reed's data appears to be a 'rationalized set', permitting computing any parameter from others listed.

It is clear that, depending on the value of χ, exponential *growth* of the neutron number density can occur. This in turn means that for a finite value of χ there is a sphere radius R_0 above which this will

This is a very familiar result for the diffusion coefficient from the transport theory of simple gases.

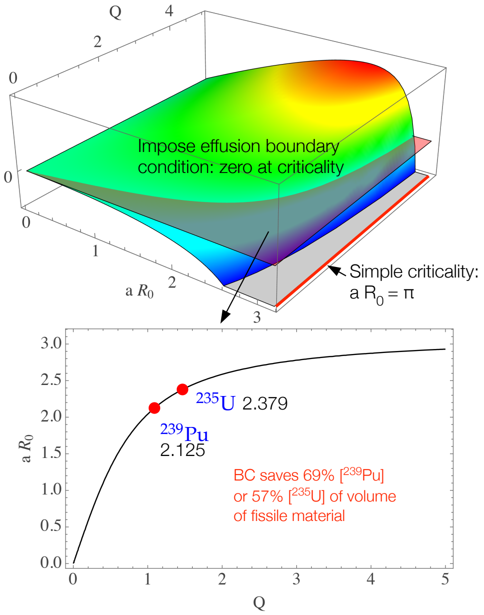

Figure 2: Criticality for a sphere of fissile material using the 'effusion' boundary condition of Eq.6. Note strong reduction of aR_0.

Table 1: Important properties for ^{235}U and ^{239}Pu relevant to neutrons produced by fission, from Fourth Edition of Reed. Radii are using the 'transport coefficient' expressions for criticality.

parameter	units	^{235}U	^{239}Pu
ν	-	2.637	3.172
n	10^{22} cm^{-3}	4.794	3.930
λ_t	cm	3.596	4.108
λ_f	cm	16.89	14.14
$\langle v \rangle$	cm/sec	1.957×10^9	1.957×10^9
D_0	cm^2/sec	2.345×10^9	2.679×10^9
χ	sec^{-1}	1.896×10^8	3.005×10^8
Q	-	1.467	1.090
R_0	cm	8.37	6.346

Table 2: Critical radius and dependence on shape and boundary conditions (BC). Z_1 is the first zero of $j_1(x)$. The effusion result is for ^{239}Pu [^{235}U].

shape	BC	$R_0\sqrt{\frac{\chi}{D_0}}$
sphere	0 density	π
sphere	effusion	2.125 (2.379)
hemisphere	0 density	$Z_1 \simeq 4.4934$

occur. The sphere volume multiplied by the density of the fissile material gives the *critical mass*. Finally, we show in Fig. 3 the advertised 'spill-over' of neutron density beyond the critical radius, in this case for ^{239}Pu.

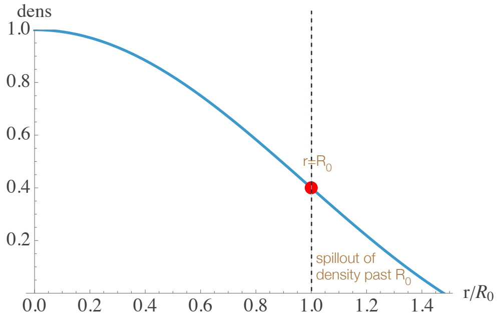

Figure 3: For ^{239}Pu, the spillover of neutron density beyond the critical radius, permitted by the effusion boundary condition.
It is a vital safety concern when storing fissile materials that sub-critical hunks not be brought near each other.

Criticality when assembling two slightly sub-critical hemispheres

This is part (b) of Mathews and Walker problem 8.2. Consider two barely stable hemispheres of fissile material assembled instantaneously to make a complete sphere and ask "What is the 'time constant' τ of the resulting exposion, as in $n(t) = n_0\, e^{t/\tau}$?".

As before, the 'chunks' of the solution for the diffusion equation in spherical polar coordinates are

$$n(r,\theta) = \sum_{\ell=0}^{\infty} \left[A_\ell\, j_\ell(ar) + B_\ell\, n_\ell(ar)\right] P_\ell(\cos\theta). \tag{13}$$

Now we *must* include angular dependence on the polar angle to permit $n\,(r = R_0)$ to be zero on the flat part of the hemisphere, which for convenience we take to be $\theta = \pi/2$. As usual, $n_\ell(r)$ diverges as $r \to 0$, so B_ℓ must be zero for all ℓ; j_ℓ and n_ℓ and the P_ℓ all oscillate faster spatially as ℓ increases. Thus the least spatially non-uniform ('lowest') solution to our diffusion equation which is zero for $\theta = \pi/2$ has $\ell = 1$ and is

$$n(r,\theta,t) = j_1(ar)\ \cos\theta\ e^{\left(\chi - D_0\, a^2\right)t} \tag{14}$$

requiring $j_1(aR_0)\ = 0$. Thus we need the same root as found in the zero-current case above, since $j_0'(x) = -j_1(x)$, or $aR_0 = Z_1 \simeq 4.49341$.

At the critical radius there is *no* time dependence and

$$\chi = D_0 a^2 = D_0 \frac{Z_1^2}{R_0^2}. \tag{15}$$

We now instantaneously assemble the full sphere by adding the lower hemisphere to the upper. What happens?

1. The sphere radius remains the same, but the *actual* spatial solution to the diffusion equation reverts to what we found for the case with $n\left(R_0\right) = 0$, namely $\ell = 0$ and a radial dependence $j_0(ar) = \sin ar/ar$, requiring for the 'lowest' solution that $ar = \pi$.

2. Since the two hemispheres were individually sub-critical, the new assembly is supercritical. The new time dependence is $e^{\chi t} \times e^{-D_0 \frac{\pi^2}{R_0^2} t}$. But we know from Eq. 15 what χt is in terms of the original hemisphere configuration. Using this, the full time dependence of the now supercritical sphere is

   $$e^{\frac{D_0}{R_0^2}\left(Z_1^2 - \pi^2\right)t} \equiv e^{\frac{t}{\tau}} \tag{16}$$

   where

   $$\tau = \frac{R_0^2}{D_0}\frac{1}{Z_1^2 - \pi^2} \simeq 0.09689\,\frac{R_0^2}{D_0} \tag{17}$$

   R_0^2/D_0 has the natural interpretation as the time taken for neutrons to diffuse throughout the sphere. The time τ is almost 10 times shorter, on the scale of nanoseconds.

The two Los Alamos fatalities due to the 'demon core' involved a ^{239}Pu sphere slightly below *criticality* and involved human error as hemispherical neutron reflectors were mis-handled, producing momentary super-critical fission.

The use of the Robin (not strictly a 'mixed') boundary condition for a critical hemisphere is discussed by Cassell and Williams [4].

Conclusions

The book *The physics of the Manhattan Project* (available in a third and fourth edition) by B. C. Reed [2] is an excellent resource. It has excellent coverage of all of the nuclear physics needed, including the liquid drop model of Bohr and Wheeler, neutron cross-sections for scattering, capture, and fission, and tabulations of transport properties for ^{235}U and ^{239}Pu.

Unfortunately, due to having been typeset without TeX and chapter-by-chapter changes in notation, the same symbol is used to indicate velocities and the number of neutrons released per fission event: be warned.

In the hands of theoretical physicists with long expertise, the Manhattan Project was successful despite the availability of only mechanical desk calculators and early punch-card machines. (See this account. The use of the diffusion equation has been supplanted by vastly more sophisticated software such as Geant4 which can track the fates of each individual neutron (including fission) and accumulate statistics using Monte Carlo. A recent review of applications to nuclear reactor kinetics and criticality (including the famous *Godiva devices*) may be found in the article by Seghour[5]. For this reason, as of 2025, there is no valid **scientific** reason for additional nuclear weapons testing–virtually any desired scenario may be accurately simulated.

References

[1] Mathews, Jon and Walker, R. L., *Mathematical Methods of Physics*, Second Edition, W. A. Benjamin, Inc (1970). This is my favorite of all terse mathematical physics books.

[2] Reed, Bruce Cameron, *The Physics of the Manhattan Project*, 4th Edition, Springer Nature Switzerland, ISBN 978-3-030-61372-3 (2021).

[3] Bickel, John H., Ph.D, *Fundamentals of Nuclear Engineering*, module 6: Neutron diffusion. Available from the U.S. Nuclear Regulatory Commission.

[4] Cassell, J.S., Williams, M.M.R., "A solution of the neutron diffusion equation for a hemisphere with mixed boundary conditions", *Annals of Nuclear Energy*, vol. 31, issue 17, pp 1987-2004, November 2004.

[5] Seghour, A., *Nuclear criticality calculations using* GEANT4, Annals of Nuclear Energy, vol. 164, p 108611 (2021).