Neutron Sources, Energy, Flux Density and Moderation
in Total-Body Neutron Activation Analysis

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There are two essential requirements for successful analysis of the elemental constituents of the body by total-body neutron activation analysis (TBNAA): homogeneity of irradiation and uniformity of counting of the induced activities. This report will consider those factors involved in achieving uniform irradiation of the target element in human subjects: namely, neutron sources, energy, flux density distribution and moderators. In addition, the radiation dose received by the patient in TBNAA, as related to the above factors, will be considered.

1. **NEUTRON SOURCES**

The potential neutron sources that can be employed for TBNAA are

a. **Constant Voltage Accelerators**: Particle energies up to 20 MeV can be obtained on Tandem Van de Graaf accelerators for protons and deuterons which can then be used for the production of neutrons with energies up to 27 MeV. Small accelerators using deuterons of a few hundred KeV and tritium targets produce large amounts of monoenergetic 14 MeV neutrons.

b. **High-Frequency Positive Ion Accelerators**: Accelerators such as the cyclotron, proton synchrotron and heavy ion linear accelerators produce neutrons with a wide range of energies. Protons above 10 MeV will produce neutrons with a wide energy range on hitting targets such as Lithium.

c. **High-Frequency Electron Accelerators**: Accelerators such as betatrons, synchrotrons and linear accelerators can produce large numbers of
neutrons from targets such as tungsten and uranium with a wide range of energies.

d. **Nuclear Reactors**: Nuclear reactors, by controlled fission, supply neutrons with energies ranging from thermal to \( \sim 15 \text{ MeV} \). These neutrons unless properly filtered are accompanied by large amounts of gamma radiation from fission products.

e. **Radioactive Neutron Sources**:

   (i) \((\alpha, n)\) sources in which an alpha emitter, such as \(^{238}\text{Pu}, \, ^{239}\text{Pu}\) or \(^{241}\text{Am}\) is mixed with the target (for example, Be), produce \( \sim 3.7 \times 10^6 \text{ n/cm}^2\text{-s} \) of neutrons with an average energy of approximately 4.5 MeV.

   (ii) \((\gamma, n)\) sources in which gamma emitters are surrounded by targets of Be or \(\text{D}_2\text{O}\) produce neutrons with energies ranging from 25 to 1000 KeV.

f. **Spontaneous Fission Neutron Sources**

   An example of this category is \(^{252}\text{Cf}\) which produces fission energy spectrum neutrons with \( \overline{E} = 2.3 \text{ MeV} \).

2. **ENERGY SPECTRA OF NEUTRON SOURCES**

Examples of the energy spectra of various neutron sources are illustrated in Fig. 1. Many of the sufficiently intense neutron sources (with the exception of 14 MeV neutron generators) produce energy spectra which are continuous and have a wide energy range.

The choice of neutrons to be employed in TBNAA depends on the element to be activated, the uniformity of the neutron flux density.
required and the radiation dose to the subject. Total body calcium, sodium and chlorine have been measured in man by the \((n, \gamma)\) reaction (1, 2, 3). For this reaction, neutrons with energies in the thermal region \((E = 0.025 \text{ eV})\) are required. However, the exponential fall-off of the thermal neutron flux density \(\theta_{\text{th}}\) in tissue \((\text{HVL} = 1.8 \text{ cm})\) precludes the direct use of an incident beam of thermal neutrons for TBNAA. Therefore, moderated fast neutrons have been used to generate thermal neutrons at depth in tissue, with tissue itself acting as an additional moderator. To date neutron energies of up to 14 MeV have been employed.

With 14 MeV neutrons from a source such as a neutron generator, both Nitrogen \(^{14}\text{N}(n,2n)^{13}\text{N}\) and Phosphorus \(^{31}\text{P}(n, \alpha)^{28}\text{Al}\) reactions occur (3, 4). \(^{13}\text{N}\) is produced at 26 times the yield of \(^{24}\text{Na}\), and \(^{28}\text{Al}(P)\) at 8 times the yield of \(^{24}\text{Na}\) (3). With lower energy neutrons such as 5 MeV neutrons, the nitrogen reaction does not occur because of its high threshold (11.3 MeV). The comparison of induced activities resulting from irradiation of the same subject with 5 and 14 MeV neutrons can be seen in Fig. 2. (Ref. 12)

Fast neutrons of energies up to 37 MeV for \textit{in-vivo} activation have been employed in mice (5). These high energy neutrons produce \(^{15}\text{O}, \; ^{13}\text{N}, \; ^{11}\text{C}\), extremely valuable parameters in medical research. Whether these high energy neutrons will be useful in activating human subjects is still in question.
RADIATION DOSE AND NEUTRON FLUX DENSITY IN HUMAN SUBJECTS

The limiting factor in TBNAA is the dose absorbed by the tissue during the procedure. Clearly, it is desirable to deliver the smallest dose possible, consistent with an accurate analysis. The energy delivered to surface tissue as a function of incident neutron energy is shown in Fig. 3. Calculations of dose absorbed by semi-infinite tissue slabs (6) or cylindrical phantoms (60 cm high, 30 cm diameter) (7) have been in general use. The recent work of Bach and Caswell (8), however, provides the most detailed data currently available. The difference between the curves for kerma and absorbed dose is presumably due to γ rays generated via the H(n, γ)D reaction and to multiply-scattered neutrons.

On the average about 50% of the energy of the neutron is lost in each collision. However, the dose per incident neutron is not strictly proportional to neutron energy, as the scattering cross-section decreases with energy at levels above ~10 KeV. Nevertheless, there is a clear increase in dose with increasing neutron energy. This renders relatively low neutron energies most desirable for TBNAA. In addition, the (n, γ) activation is directly proportional to thermal neutron flux density ($\phi_{th}$). Thus, the efficiency with which $\phi_{th}$ is generated is of importance. The distribution of the fast neutron dose in cylindrical phantoms is shown in Fig. 4 (7). As can be seen, there is a dramatic increase in penetration with increasing incident neutron energy. This increase results mainly from the reduction in scattering cross-section, from a slight forward peaking in the scatter-
ing at higher energies, and from an increase in the number of collisions necessary for thermalization. The increased penetration also results in a significant increase in the number of neutrons escaping from an anthropomorphic phantom. Thus, fewer thermal neutrons are generated per incident fast neutron. The overall effect is an increase in the efficiency of \( \phi_{th} \) production at lower incident neutron energies.

The distribution of \( \phi_{th} \) in tissue is a sensitive function of phantom size, as well as incident beam size and uniformity. Therefore, intercomparison between differing geometries are difficult to effect. However, two sets of experiments which present \( \phi_{th} \) distributions in tissue for various incident neutron energies are available (9, 10). Smith and Boot (9) utilized accelerator-produced neutrons at energies of 0.13, 0.5, 1.0, 2.5 and 14.1 MeV and a Po-Be source (\( \bar{E} \sim 4.3 \) MeV). Aceto and Churchill (10) employed Sb-Be (photoneutrons), Po-Li, Po (mock fission) and PuBe neutron sources, with \( \bar{E} = 0.030, 0.46, 1.6 \) and 4.2 MeV, respectively. By normalizing the two sets of experiments (at 4.2 MeV), one obtains the relative efficiency with which thermal neutrons are generated as a function of incident neutron energy. These results, expressed as the ratio of peak thermal neutron flux density per incident neutron relative to the ratio obtained at 14 MeV, are shown in Fig. 5.

The ordinates express the efficiency with which thermal neutrons are generated by various incident neutron energies relative to 14 MeV neutrons (\( \phi_{th} \) advantage factor). It is apparent that, (as with dose
an advantage is obtained with the use of low energy neutrons. If a similar advantage factor curve is constructed from Fig. 3 (i.e., the ratio of energy delivered to tissue (14 MeV)/energy delivered to tissue (E)), the product gives the total advantage to be gained relative to 14 MeV neutrons, (Fig. 6). The product advantage factor is seen to rise significantly and monotonically with decreasing incident neutron energy.

If it is desired to measure absolute levels of whole-body Ca, it is necessary to have fairly uniform $\phi_{th}$ distributions. This results from the fact that the target (Ca) distribution is variable as well as unknown. From Fig. 7 it is evident that the $\phi_{th}$ peak shifts to shallower depths as incident neutron energy decreases (10). Since it is desirable to measure Ca in volumes up to $\sim 25$ cm in thickness, it is clear that very low neutron energies will not be usable, since poor penetration precludes a uniform $\phi_{th}$ distribution at depth. Hence, a compromise has to be made between the advantage gained through the use of low energy neutrons and uniformity of $\phi_{th}$ distribution which increases with neutron energy.

To date, uniformity of flux density of thermal neutrons through the human subject has not been achieved. Various investigators have coped with this problem of achieving uniform fluence in different ways (11). By suitable selection and positioning of the moderator, a highly uniform flux density can be obtained through the body including the first few centimeters of tissue. This is of importance since a large portion of the skeleton (Ca) lies within a few centimeters
of the surface. For the activation analysis of homogeneously distributed target elements, the high entrance flux is not as critical.

The comparative flux densities from some of the neutron sources are shown in Fig. 8. For these measurements, single sources (no reflector) were used, and the results "cross over", to yield bi-lateral curves. As seen from the figure, with bi-lateral irradiation, the most uniform flux density, as measured in a tissue-equivalent phantom, is obtained with moderated 14 MeV neutrons. The \( \Phi_{th} \) distribution produced by \(^{252}\text{Cf} \quad (\bar{E} = 2.3 \text{ MeV}) \) indicates too great a range (± 25.5%) to be useful for absolute Ca analysis. Alpha-n sources such as \(^{238}\text{Pu-Be} \) produce the characteristic bi-modal energy spectrum, with \( \bar{E} = 4.2 \text{ MeV} \), and a reasonable uniform \( \Phi_{th} \) distribution (see Fig. 1). This reasonably uniform \( \Phi_{th} \) distribution coupled with the advantage gained through the use of a relatively low energy incident neutron (advantage factor = 2.1 relative to 14 MeV neutrons, see Fig. 6) makes the use of \(^{238}\text{Pu-Be} \) sources attractive for TBNAA. An additional benefit results from the ease of operation and reproducibility inherent in constant-output radioactive neutron sources (12).

A number of techniques based upon the scattering properties of neutrons can be used to enhance the output and characteristics of sources for TBNAA. Fig. 9 illustrates the improvement obtained through the use of a point source at 150 cm SSD vs the source at 50 cm SSD. The results (corrected for inverse square variation) indicate the advantage of "broad-beam" conditions, where the peak \( \Phi_{th} \) per incident fast neutron was increased by 1.55 relative to the point
source at 50 cm.

Also, the use of a high Z reflector (constructed from material such as bismuth, with a low reaction cross-section) will back-scatter fast neutrons, thus "broadening" the beam, as well as reducing the amount of radioactivity needed in the neutron source. Fig. 10 shows that the addition of a 3-inch Bi reflector (reflector occupied ≤ 2π of the available 4π steradians) increased the effective output of the source by ~1.4 (peak position). Thus, the use of 238Pu-Be neutrons for TBNAA in a "broad-beam" geometry with a Bi reflector gives more counts per unit absorbed dose and thus yields an overall advantage of 3.6 relative to 14 MeV neutrons (12).

4. SUMMARY

The factors involved in achieving uniform irradiation of the target element in total-body neutron activation (TBNAA) are considered. The neutron sources that can be employed are accelerators of various types, nuclear reactors, and radioactive sources (α, n), as well as spontaneous fission sources (252Cf).

The choice of neutrons to be employed in TBNAA depends on the element to be activated, the uniformity of neutron flux density required and the radiation dose to the subject. The limiting factor in TBNAA is the dose absorbed by the tissues during the procedure. The dose per incident neutron is proportional to the energy. Further, the activation produced by the n,γ reaction is proportional to the thermal flux density (φth). The φth produced is inversely proportional to the neutron energy. Thus the use of low energy neutrons still further increases the advantage compared with 14 MeV neutrons. However, a
compromise must be made between this advantage and the desired uniformity of $\phi_{th}$ distribution which increases with neutron energy.

Various degrees of uniformity of flux density of thermal neutrons through the body of the human subject have been achieved by investigators by the selection of appropriate moderators which thermalize the fast neutrons prior to their entrance into the body. A consideration of all the above factors indicates that $\alpha, n$ neutron sources, such as $^{238}$Pu, Be, have an advantage of $\sim 3.6$ relative to 14 MeV neutrons in TBNAA.
REFERENCES


10. ACETO, H. and CHURCHILL, B.W., Neutron depth dose from (α, n) and (γ, n) sources in a tissue-equivalent phantom, Lawrence Radiation Lab. Report, UCRL 10267, 1963.


FIGURES

Fig. 1  Energy Spectra of Neutron Sources
A. Pu,Be (α,n) - Stewart, 1955)
B. Accelerator Neutron Spectra
(A) 12 MeV protons on thick Be targets
(B) 6 MeV protons on thin Be targets
(C) 22 MeV protons on thick Be targets
(Tochilin & Kohler, 1958; Gugelot, 1951; Sheppard and Darden, 1953)
C. Accelerator Deuterons on Be Target (Cohen and Falk, 1951;
Tochilin and Kohler, 1958)
D. Spontaneous Fission 252Cf, 235U and 244Cm. (Stoddard, 1964).

Fig. 2  Comparison of Spectra Following Irradiation of Same Subject with 14 and 4.5 MeV Neutrons.

Fig. 3  Energy Delivered to Tissue as Function of Incident Neutron Energy.

Fig. 4  Distribution of Fast Neutrons in Cylindrical Phantom.

Fig. 5  Ratio of Peak Thermal Neutron Flux Density per Incident Neutron Relative to Ratio Obtained at 14 MeV.

Fig. 6  Total Advantage to be Gained Relative to 14 MeV Neutrons. Product of Dose Advantage Factor x ϑ th Advantage Factor.

Fig. 7  Peak Thermal Neutron Flux Related to Depth as a Function of Neutron Energy.

Fig. 8  Depth Flux Distribution for Bi-lateral Irradiation of Tissue-Equivalent Phantom with Various Neutron Sources.

Fig. 9  Comparison of "Broad-beam" Neutron Source with Point Source.

Fig. 10  Effect of Bismuth Reflector on Neutron Flux.
FIGURE 2

- $^{13}\text{N}$ (0.51 MeV)
- $^{38}\text{Cl}$ (1.64 MeV)
- $^{24}\text{Na}$ (1.37 MeV)
- $^{28}\text{Al}$ (1.78 MeV)
- $^{38}\text{Cl}$ (2.16 MeV)
- $^{24}\text{Na}$ (2.76 MeV, 49Ca (3.1 MeV)

- 14 MeV NEUTRONS
- Pu, Be NEUTRONS (~5 MeV)

Count/15 Minutes

Channels (33 keV/Channel)

Neg. # 1-55-72
ENERGY DELIVERED TO TISSUE (STANDARD MAN)
vs
NEUTRON ENERGY

- ○ KERMA FOR TISSUE IN AIR (BACH AND CASWELL)
- x MAXIMUM ABSORBED DOSE TO CYLINDER, 
  HT.= 60cm, D=30cm (NCRP NO.38)
- □ MAXIMUM ABSORBED DOSE TO SEMI-INFINITE SLAB (USNBS NO.63)

ERGS PER GRAM PER 10^-7 n/cm^2

NEUTRON ENERGY IN MeV

Figure 3
RATIO OF PEAK THERMAL NEUTRON FLUX DENSITY PER INCIDENT FAST NEUTRON RELATIVE TO THAT VALUE AT 14 MeV

$\left( \frac{\phi_{th}}{\phi_F} \right) / \left( \frac{\phi_{th}}{\phi_F} (14 \text{ MeV}) \right)$

NEUTRON ENERGY IN MeV

FIGURE 5
PRODUCT OF DOSE ADVANTAGE FACTOR 
\( \times \phi_{th} \) ADVANTAGE FACTOR

FIGURE 6
FIGURE 7

Thermal-neutron flux peak depth into phantom as a function of incident neutron energy.
(From Aceto & Churchill ref. 10)
DEPTH FLUX FOR
BILATERAL IRRADIATION
OF TISSUE EQUIVALENT
PHANTOM

Na$_2$CO$_3$ FOILS IN
TISSUE EQUIVALENT
LIQUID

- Pu Be (5 Ci) 50 cm SSD
- Am Be (50 Ci) 50 cm SSD
- C$^{252}$ 50 cm SSD

VARIATION IN
FLUX

±6.9%
±10.3%
±25.5%

DEPTH IN T.E. PHANTOM, cm

PERCENT DEPTH FLUX

120
110
100
90
80
70
60
50
40
30
20
10

3 cm BUILD-UP

5 cm BUILD-UP

14 MeV TEXAS NUCLEAR
NEUTRON GENERATOR
130 cm SSD

FIGURE 8
THERMAL NEUTRON FLUX DENSITY
RELATIVE TO PEAK VALUE AT 50 cm SSD

\[ I = 50 \text{ Ci Am Be SOURCE}, 150 \text{ cm SSD} \]
\[ \circ = 50 \text{ Ci Am Be SOURCE}, 50 \text{ cm SSD} \]

FIGURE 9
RELATIVE THERMAL NEUTRON FLUX DENSITY

FIGURE 10

DEPTH IN cm

O = 50 Ci Am Be SOURCE, 50 cm SSD, NO REFLECTOR
X = 50 Ci Am Be SOURCE, 50 cm SSD, 3 in. Bi REFLECTOR

THERMAL NEUTRON FLUX DENSITY RELATIVE TO PEAK VALUE, WITH NO REFLECTOR