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COMPARISON OF TECHNIQUES

for the

TOTAL-BODY NEUTRON ACTIVATION ANALYSIS OF CALCIUM IN MAN

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An accurate measure of the skeletal mass in patients with bone demineralizing diseases is of obvious importance. Radiographic techniques are generally inadequate for detection of subtle changes in bone which result from disease. The technique employing photon absorption, while precise, yields information only on localized bones of the extremities. It is equally difficult to extrapolate to the whole skeleton from a study of bone biopsy samples. There is general agreement that total-body neutron activation is the only currently available technique by which a precise and accurate measure of the total bone mass may be made.

This report will review the present state of the art in the measurement of total body calcium. Battye (1) in 1967 demon-

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strated clearly the feasibility of measuring total body Ca by the reaction $^{48}\text{Ca}(n, \gamma)^{49}\text{Ca}$. Partially-moderated 14 MeV neutrons were used along with a bi-lateral exposure. Three groups of investigators have applied this technique for measuring total-body calcium in man: the Univ. of Birmingham group, the University of Washington group, and the Brookhaven group.

Since the reports of the results of all of these groups have been published, this report will develop a comparison of the salient features of the three approaches to the central problem. General conclusions will be drawn, and these can provide a basis for discussion of current work in progress. Moreover, a review of the problems encountered by the investigators and the measures taken to counter them may point the way to possible improvements in techniques for measuring total body calcium.

I. Neutron Sources and Patient Exposure Geometry

Of necessity, all of the initial sources of neutrons employed were sources designed for research in physics. Thus, Chamberlain (2,3) and Nelp and Palmer (4, 5) have employed the cyclotron (60") produced neutrons with peak energies of 3.5 and 8 MeV, respectively. The spectra are continuous, and have wide energy ranges, (see Table 1). Cohn (6,7) originally used 14 MeV neutrons from a D,T neutron generator, but has, more recently, employed α, n neutron sources (8).

In Chamberlain's technique, the patient is irradiated while lying on his side in a polypropylene chamber (0.9 x 0.9 x 2 m) to provide moderation of incident fast neutrons. In Nelp's technique, the patient is irradiated upright, with the extremities in lucite containers of water, and the head encased in a plastic helmet for moderation. Cohn employed 14 MeV ^{neutrons}, with the patient standing upright, supported by crutches, and positioned tightly between two large sheets of 3.8 cm polyethylene moderator. In all three techniques, the irradiation is performed first with the patient facing the target, and then with the patient facing in the opposite direction. None of these procedures are particularly physically comfortable for the patient.

It was for this reason that the BNL group designed a portable α, n source, installed in the hospital, which permits a simultaneous bi-lateral exposure of a patient resting supine on a bed (8). The source of neutrons in this procedure is a bank of 14 encapsulated 50 Ci sources of $^{238}\text{Pu, Be}$ ($\bar{E} \sim 4.2 \text{ MeV}$), positioned above and below the length of the patient.

The success of the total body activation analysis procedure for the measurement of total-body calcium content (particularly absolute levels) depends essentially on meeting two requirements, (and in some cases, three).

- A. A uniform thermal neutron distribution must be obtained in order to expose uniformly the target element in the

body to a standardized quantity of neutrons.

- B. A quantitative measure of the induced ^{49}Ca activity in the body must be obtained with a whole-body counter having uniform counting sensitivity.
- C. A third requirement, important only when 14 MeV neutrons are employed, is the correction for interfering radio-nuclides produced from different target elements present. The reaction $^{38}\text{Cl}(n,p)^{37}\text{S}$ is of prime importance, since ^{37}S has the same energy as ^{49}Ca .

II. Uniformity of Thermal Neutron Depth Flux Density in the Irradiated Subject

The problem of obtaining uniform depth flux density of thermal neutrons in man is complicated by his non-uniform shape (9). In the measurement of Ca, which is concentrated almost entirely in the skeleton, the problem of obtaining uniform flux to the target is even more difficult than in the case of more homogeneously distributed elements such as N, Na and Cl. While most of the skeleton lies within a few centimeters of the skin, there are large differences in the amount of overlying tissue which also serves to moderate the incident neutrons. To achieve any degree of uniformity of flux, all investigators have found it necessary to utilize fast neutrons with moderators and a bilateral exposure. Obviously, in order that absolute measurements can be made, the degree of uniformity required is much

higher than that required for relative measurements. It is most desirable to activate as large an amount of the skeleton as possible. Besides high sensitivity, a high degree of precision, i.e., reproducibility, is a necessity, if meaningful relative measurements are to be obtained.

In all three techniques for total body Cs measurement, the depth flux density profiles of thermal neutrons were estimated with the use of cubic phantoms of water (1,5,6) or hydrogenous plastic (3), of approximately the thickness of the human body positioned in the exposure geometry.

Chamberlain, using cyclotron-produced neutrons ($\bar{E} \sim 3.5$ MeV) with a 20 cm thick polyethylene phantom, found a peak flux at 3 cm from the surface of the phantom when 1.5 cm of polypropylene moderator was used (3). The uniformity of neutron flux was $\pm 27\%$ (3).

Nelp (5) obtained a uniformity of neutron flux density in a cubic water phantom of 25.4 cm depth with no moderator of $\pm 18\%$. The maximum flux occurred at 4.5 cm from the surface. To assess better the uniformity of flux in the human body, Mn capsules ($1/2''$ by $1''$) were placed at $1''$ intervals throughout the chest, thorax, extremities and head of a cadaver. The variation in the activation of these capsules was $\pm 20\%$ of the median value. With the Mn capsules located in individual bones of a cadaver, the standard deviation was 5.6% and the range of

activation flux uniformity about the median value was $\pm 12\%$.

Cohn, using 14 MeV neutrons and a 5 cm moderator of paraffin placed up against the front and back of the phantom, found that the uniformity of neutron flux over 23 cm was $\pm 5\%$ (6), see Fig 1. Battye (1) reported uniformity of flux was $\pm 3.5\%$, with 3.5 cm of a T.E. material and a field separation of 25 cm.

With the $^{238}\text{Pu,Be}$ neutrons, Cohn found that the uniformity in a cubic water phantom of 28 cm thickness and 2.5 cm of moderator was $\pm 15\%$. With 1.9 cm polyethylene moderator closely fitted plus an assumed 1 cm of tissue serving as additional moderator, a uniformity of $\pm 8\%$ was obtained in an Alderson phantom (8). Part of this uniformity is due to the use of a 3" Bi reflector lining the irradiation cell, which greatly increased the non-degraded reflectance of the generated neutrons (9).

III. Moderator

The thickness of the hydrogenous moderator and its position in relation to the body is critical in obtaining a uniform flux through the body. Obtaining the maximum build-up of the incident neutron flux in the moderator must be balanced against the decreased penetration of the neutrons in the body.

A second factor that affects the uniformity of the neutron flux is the position of the moderator in relation to the body. With an air-space between the moderator and the skin (MSD), there is a divergence of the neutron beam, and a secondary

build-up when the neutrons hit the body.

Considerable differences in moderators, both in their thickness and in their positioning, exist among the three studies. Chamberlain employs a 1.3 cm thick polypropylene moderator in the form of a box 0.3 x 0.3 x 2 m, in which the patient is placed. He reports, however, that the divergence of thermal neutrons which results from the large MSD is largely counterbalanced by the presence of neutrons moderated in other parts of the box. In the technique employed by Nelp, a variety of moderators are used, ranging from a 6" thick lucite boot filled with water for the limbs, to a $4\frac{1}{2}$ " thick lucite cylinder filled with H_2O for the arms, to a $3\frac{1}{4}$ " thick lucite helmet for the head and neck, to no moderator over the chest and the abdomen,(5). Nelp points out that over 50% of skeletal Ca resides in the extremities and the skull.) Further, the subject is surrounded on the sides, top and back by 6" of plywood to shield him from neutrons which are scattered toward the patient from other surfaces of the room.

Cohn attempted to minimize the error in the positioning of the moderator with the 14 MeV neutrons by positioning the patient tightly between two sheets of 3.8 cm thick polyethylene. There remains, however, air gaps between the moderator and the skin in this technique, due to the shape of the body. In particular, the largest air gaps occur with the extremities. With the $^{238}\text{Pu,Be}$ neutron facility, an attempt was made to fit the

moderator as closely to the patient as possible. The patient rests supine on a sheet of 1.9 cm thick polyethylene, (see Fig. 2), and is covered with a number of form-fitting sections of moderator to maximize the uniformity of the thermal fluence through the body (8). The choice of section of moderator and position of the moderator is recorded, and both can be exactly reproduced for subsequent activations. The closely-fitting moderator also serves to fix the position of the patient.

A solid plastic tissue-equivalent anthropomorphic phantom (Alderson) containing a human skeleton was used to establish empirically the optimal thickness and position of the moderator. The relative ^{49}Ca count obtained with the various moderators or with no moderator is shown in Fig. 3. Clearly, there is an optimum thickness of moderator in terms of achieving the optimal activation of Ca in the body. The absence of moderator, or the presence of too much moderator, or the inclusion of as little as a 6" air gap between the moderator and the skin, all serve to reduce the level of ^{49}Ca produced. With no moderator, the Ca situated close to the surface of the body is underactivated. With too thick a moderator, or with too low an energy of incident neutrons, the peak flux will occur near the skin surface, and again underactivate the skeletal Ca.

Undoubtedly, these differences are minimized in this situation due to the high degree of reflectance achieved by lining the irradiation cell with 3" of bismuth. Clearly, the selection of moderator to obtain the maximum activation of skeletal Ca is

an empirical procedure for neutrons of energy < 14 MeV. However, in each of the irradiation-moderator techniques described above, the precision of reproducibility of the activation is very high, 1 - 3%; the accuracy is not nearly so good.

IV. Measurement of Induced ^{49}Ca Activity in the Body

Since the energy of ^{49}Ca is high (3.1 MeV) the attenuation and geometry correction is not as large as for the measurement of ^{13}N , $^{28}\text{Al}(\text{P})$, and Na and Cl, but it cannot be ignored. In the Brookhaven technique, the whole-body counter-computer facility corrects for both geometry and attenuation effects for the individual patient being analyzed (10).

The counting efficiency varies with the volume of NaI detector employed; and it ranges from 4689 cc in Chamberlain's procedure to 50,000 cc in the Brookhaven counter, (see Table 1). The BNL counter also has a further advantage in that it counts the entire body simultaneously, while Nelp's counter scans the body with an annular array of four $4 \times 9\text{-}3/8$ " NaI detectors. These detectors move across the body at an exponential rate, synchronized to the $t^{1/2}$ of ^{49}Ca . Chamberlain employs four $5" \times 4"$ detectors in a fixed geometry (3). If the counts in the ^{49}Ca photopeak are divided by the rem dose to the patient, the ratio for Chamberlain, Nelp and Cohn (14 MeV) and Cohn (α, n) neutrons are 1:1.8:2.7:7.7 respectively, (see Table 1).

V. Absolute Measurement of Calcium

In both the techniques of Chamberlain and Nelp, absolute

measurement is not attempted. Chamberlain related the induced ^{49}Ca to the simultaneous ^{24}Na induced. This has the advantage of eliminating uncertainties in the neutron flux measurement, but has the disadvantage in that total body sodium itself is a variable. Since it has been estimated that 30% of Na is located in bone, this is a particularly questionable technique in the study of patients with bone-wasting diseases. The variability of Na content can be decreased somewhat by simultaneous measurement of exchangeable Na with a tracer in each patient analyzed.

Nelp has chosen to express the induced ^{49}Ca in terms of four 1.2 liter Lexan containers, each containing 1600 g of Ca, and mounted on each side of the exposed subject. These are stirred to correct for the non-uniform depth flux and then counted in a whole-body counter after the patient.

Cohn has attempted to estimate the absolute grams of Ca by a series of experiments employing an Alderson phantom. Since an Alderson phantom containing homogeneously distributed Ca targets does not simulate too well the Ca distribution in the skeleton, the following experimental approach was employed to determine absolute level. A calibration factor which relates the amount of target, neutron flux and induced ^{49}Ca activity was first determined, (see Table 2).

$$CF = \frac{m \phi_{th}}{A}$$

m = mass of Ca

ϕ_{th} = average thermal neutron flux, $n/\text{cm}^2\text{-sec}$

A = corrected counts/15 min.
in 3.1 MeV photopeak of
 ^{49}Ca corrected to $t = 0$

Measured targets of Ca were irradiated in a standardized thermal beam of the BNL reactor. The average thermal flux was measured by activated Au foils surrounding the target. The activated Ca target was then dissolved in water and distributed homogeneously in an Alderson phantom. The absolute activity of ^{49}Ca was measured in the 54-detector whole-body counter (corrected for geometry and attenuation), (see Table 3).

After the CF was calculated, measured amounts of Ca were homogeneously distributed in the phantom, and the phantom was irradiated in either the 14 MeV facility or the $^{238}\text{Pu,Be}$ facility and subsequently counted in the whole-body counter. The depth flux density was $\pm 5\%$ and $\pm 8\%$ for the two neutron sources, respectively; hence, the probability that the entire skeleton is activated is $\pm 5\%$ and $\pm 8\%$. The calibration factors calculated by these two procedures were the same.

VI. Dose Absorbed by the Patient

The dose to the patient in the techniques reviewed here (generally measured with tissue equivalent ionization chambers) is shown in Table 1. The dose ranges from 0.277 rem for $^{238}\text{Pu,Be}$ neutrons to approximately 2 rem (Nelp and Chamberlain). Comparison of the measured doses with theoretical calculations have also been carried out for 14 MeV neutrons and for neutrons from various α, n sources, (see Table 4). (Data on this point from Nelp and Chamberlain are insufficient).

It is clear that the ~ 4.5 MeV neutrons have a considerable

advantage over the 14 MeV neutrons in terms of reduced dose to the patient required to achieve the same activation. The factors responsible for this reduction are:

1. ratio of dose to number of incident neutrons
2. ratio of thermal neutrons per incident neutron
3. effect of simultaneous bi-lateral irradiation
4. effect of "broad-beam" source on ϕ_{th}/ϕ_f ratio
5. greater reflectance of incident flux due to Bi reflection, (Albedo effect)
6. reduction of transport time from irradiation to counter

VII. Conclusion

There are two requirements essential for the measurement of the body Ca content by total body neutron activation: a uniform thermal neutron distribution through the body, and uniform counting sensitivity of the whole-body counter. Of the two, the first is the more difficult to attain. Clearly, the achievement of the highest degree of uniformity of thermal neutrons is an art, and there is considerable space for improvement of technique. The uniformity obtained is a function of the energy of the incident neutron energy (9) and the thickness and positioning of the moderator. The best results are undoubtedly obtained with moderated 14 MeV neutrons. Adequate uniformity, at least for relative measurements, has, however, been obtained with lower energy cyclotron-produced neutrons.

The uniformity of counting of the induced activity is only critical if the absolute measurement of induced ^{49}Ca is desired.

Besides the reduced dose to the patient, there are numerous advantages derived from the use of α, n neutrons over neutrons produced by cyclotrons or neutron generators. The irradiation geometry is substantially improved, and significantly greater stability in neutron output is achieved, and hence the precision of the technique is enhanced. The operation of α, n neutron sources is greatly simplified, and a trained operator is not required, as is the case with neutron generators and cyclotrons. Finally, the design of such a facility for medical applications, permitting it to be located in a hospital environment, is a significant advantage to the patient, as well as to the investigator.

It is clear that if total-body neutron activation analysis is eventually to become a part of the armamentarium of the medical profession, the most likely direction of development will be in the application of "portable" α, n neutron sources.

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ILLUSTRATIONS

- Fig. 1** Depth flux for bi-lateral irradiation of tissue-equivalent phantom. Depth flux curves for 14 MeV neutrons as well as those for Pu,Be, Am,Be and ^{252}Cf are shown.
- Fig. 2** The polyethylene moderator surrounding patient prior to irradiation exposure. The moderator is sectional and is fit to conform to the shape of the individual.
- Fig. 3** Effect of varying moderator thickness and position in activation with $^{238}\text{Pu,Be}$ neutrons.

FIGURE 1

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FIGURE 2

**EFFECT OF VARYING THICKNESS AND
POSITION OF POLYETHYLENE MODERATOR ON
ACTIVATION OF SKELETAL CALCIUM FOLLOWING
IRRADIATION WITH $^{238}\text{Pu}, \text{Be}$ NEUTRONS.**

TABLE I

SUMMARY OF NEUTRON SOURCES, FLUX AND LEVELS OF INDUCED
⁴⁹Ca ACTIVITY IN VARIOUS TOTAL-BODY NEUTRON ACTIVATION TECHNIQUES

Experimental Groups	Neutron Source	Energy MeV	Incident fast neutron flux, n/cm ² -sec	Uniformity of thermal flux in body (0/0)	Whole-body Counter, detector volume	Induced ⁴⁹ Ca Activity c/t	Precision for measurement of Ca (%)	Patient absorbed dose			⁴⁹ Ca activity rem
								n rad	y rad	total rem	
University of Birmingham	Cyclotron ⁷ Li(p,n) ⁷ Be	0.1-8 peak = 3.5	3 × 10 ⁵ at 2 m	± 15 cubic 20 cm phantom with 1.5 cm moderator.	4 (5" × 4") 4689 cc	5800/17min at 7 min	± 3.7	0.098 .080 .178	0.05	1.83	3164
University of Washington	Cyclotron ⁹ Be(d,n) ¹⁰ B	4-12 peak = 8	N.A. at 3.7 m	± 5.6 in cadaver with variable moderators	4 (9-3/8 × 4") 18142 cc (no shielding)	12000/12.5 min at 4 min	± 1.3	0.20	0.10	2.10	5714
Brookhaven	Neutron generator	14	4.20 × 10 ⁶ at 1.5 m	± 5 cubic 23 cm phantom with 3.8 cm moderator	54 (6" × 2") 49948 cc	5500/15 min at 6 m	± 1.7	0.063	0.007	0.637	8634
Brookhaven	²³⁸ Pu, Be 700 Ci ⁹ Be(α,n) ¹² C	~4.5	3.09 × 10 ⁶ at 0.3 m	± 8 in Alderson phantom with 2.9 cm moderator	54 (6" × 2") 49948 cc	6800/15 min at 3 m	± 1.0	0.026	0.02	0.28	26283

TABLE 2

Calculation of calibration factor for estimating total-body calcium

Run	Mass of calcium (m) (mg)	Flux density (ϕ_{th}) (10^{10} n cm $^{-2}$ sec $^{-1}$)	Activity (for 5-min irradi.)		Calibration factor (CF) (10^3)
			Counts/15 min point source	Counts/15 min (A) phantom	
1	42.010	6.823	323,855	228,495	12.55
2	41.675	9.780	470,788	333,250	12.23
3	42.300	10.022	489,076	354,127	11.97
					<u>12.25</u>

$$CF = m \cdot \phi_{th} / A$$

where: m = mass of Ca (g)

ϕ_{th} = thermal neutron flux density (n cm $^{-2}$ sec $^{-1}$)

A = corrected counts/15 min in the 3.1-MeV photopeak of ^{49}Ca , corrected to $t = 0$

TABLE 3. Experimental determination of calcium in Alderson phantom by neutron irradiation

Run	Time of irradiation (min)	ϕ_{th} (10^8 n cm $^{-2}$ sec $^{-1}$)	Target in phantom			counts/15 min* at $t = 0$	Calc. Ca (g)	Error (%)
			Ca (g)	Na (g)	Cl (g)			
1	8.9	3.790	348.4	0	0	24438 23447 <u>23943</u>	333.3	-4.33
2	8.9	4.066	348.4	0	0	27433 27029 26738 <u>27066</u>	351.3	0.83
3	5	4.573	1000.0	105	105	55767 55354 54628 <u>55270</u>	907.4	-2.26
4	5	4.553	1000.0	105	105	56983 54669 <u>55850</u>	1002.2	0.22
5	5	4.476	1000.0	105	105	53090	969.1	-3.1
							$\bar{x}$	-1.73

* Corrected for geometry and attenuation. CF for 8.9 min irradiation = 5.278×10^3 . CF for 5.0 min irradiation = 8.170×10^3 .

TABLE 4

Estimation of Absorbed Dose for Bilateral Irradiation
with Various Neutron Sources

	$\phi_{th}/\phi_f(1)$	ϕ_f in Units of 10^4 $n \cdot cm^{-2} \cdot sec^{-1}$	Kerma Rate in Units of 10^{-2} ergs $gm^{-1} \cdot sec^{-1}$	Rads $hr^{-1}(2)$
<u>14-MeV Neutrons</u>				
Texas-Nuclear Generator	1.08	7.00(3)	4.67(4)	1.68
Smith & Boot (Ref. 6)	1.21	6.33	4.22	1.52
<u>Portable (α, n) Neutrons Source</u>				
AmBe	1.19	0.817(5)	0.343(6)	0.123
PuBe	1.66	0.589	0.247	0.089
PoBe	1.36	0.716	0.300	0.108

(1) ϕ_{th}/ϕ_f = incident fast neutron flux density/thermal neutron flux density.

(2) Experimentally, the contribution to the absorbed dose from multiple scattered neutrons was not found to be significant; therefore, the kerma can be equated with the absorbed dose to surface tissue.

(3) Based on the minimum ϕ_{th} required to produce a whole-body count of 5000 cts/15 min in the 3.0-MeV photopeak at 5 min postirradiation. Irradiation time of 5.0 min. This value was reduced by a factor of 1.20 to account for the reduction in absorbed dose resulting from the polyethylene moderator.

(4) Kerma rate calculated using a dose conversion factor of 6.67×10^{-9} rads $n^{-1} \cdot cm^{-2}$ (Ref. 7)

(5) Based on the minimum ϕ_{th} to produce a whole-body count of 5000 cts/15 min at 2 min post-irradiation. Irradiation time = 17.6 min. This value is corrected for simultaneous "broad-beam" bilateral irradiation with a polyethylene moderator.

(6) Kerma rate calculated using a dose conversion factor obtained from mean energy derived from AmBe spectrum (4.2×10^{-7} ergs $gm^{-1} \cdot n^{-1} \cdot cm^{-2}$). Similar energy distribution assumed for PuBe and PoBe. Kerma rate can also be calculated using the dose conversion factor of 5×10^{-7} ergs $g^{-1} \cdot n^{-1} \cdot cm^{-2}$ (Snyder & Neufeld, NBS 63).