Reports

Gamma-Emitting Radionuclides in Newborns, Infants, and Children

Abstract. *During the period April 1962 to April 1963, 28 newborn infants, 7 stillborns, and 21 children from 4 to 15 years of age were examined for emission of gamma activity in a low-background, high-sensitivity, total-body counting facility. The 21 children contained traces of combined Zr^{95} and Nb^{95}, $Ru^{103, 106}$, and Cs^{137} in addition to the normal K^{40}. The greatest concentration of Cs^{137} observed was 80 picocuries per kilogram of body weight, or about 4 percent of the natural K^{40} radioactivity. Four of the seven stillborns, each counted for 10 hours, showed K^{40} and traces of Zr^{95}, Nb^{95}, and $Ru^{103, 106}$. The ratios of Zr^{95} to Nb^{95} were lower in the stillborns than in their placentas which suggested placental discrimination against Zr^{95}. The ratios of Cs^{137} to K^{40} were lower in infants than in children or adults. Thyroid glands obtained at autopsy from 24 infants during April to September 1962 contained no detectable radioiodine (less than 30 pc).*

Measurements of the quantities of several gamma-emitting radioactive nuclides in the bodies of human beings have been reported for population samples of several nations (*1*). These data permit rough estimates of the body burdens currently existing in adult populations, but not in populations of infants or young children. This report presents results of such measurements made between April 1962 and April 1963 on a number of stillborn and newborn infants and children from the environs of Los Angeles, California. In addition, assays for radioactive iodine were made of fetal and neonatal thyroid glands obtained at autopsy in the period May to September 1962 during which atmospheric nuclear tests were being conducted. The measurements were made inside a steel cubicle (2.4 × 2.4 × 2.4 m) with 15-cm thick walls; scintillation detectors with a large sodium iodide crystal and multichannel pulse-height analysis (*2*) were used.

For measurements on children, four sodium iodide crystal detectors, each 12.5 cm in diameter × 10 cm thick were arranged, two above and two below a plastic cot where each child was placed in a supine position during the counting time of 30 to 45 minutes. Twenty-one children ranging from 4 to 15 years of age were examined. In each instance the total absorption peaks for the primary gamma photons from naturally occuring K^{40} and fission product Cs^{137} (1.46 and 0.66 Mev, respectively) were prominent. Characteristic peaks (full-energy absorption) ascribable to combined Zr^{95} and Nb^{95} and to $Ru^{103, 106}$ were also detected. In several instances where these peak heights were about twice the K^{40} peak, special scrubbing and shampooing reduced the observed Zr^{95} and $Ru^{103, 106}$ emission considerably, but did not eliminate it—which suggested that a portion of this activity was within the body. In six children very low peaks appeared in the 0.32 to 0.39 Mev band. Whether these were due to I^{131} (0.36 Mev) or La^{140} (0.33 Mev) could not be determined at this low level, since the other major characteristic peaks of La^{140} (and its parent Ba^{140}) were masked by the prominent peaks of Zr^{95} and $Ru^{103, 106}$ which nearly coincide. The other La^{140} peak at 1.60 Mev has a lower relative height than the 0.33 Mev peak and was not detectable.

The K^{40} concentrations ranged from 1800 to 2600 pc (1 pc = 10^{-12} curie) per kilogram of body weight which corresponds to 2.0 to 2.9 g of potassium per kilogram of body weight. The greatest concentration of Cs^{137} observed was 83 pc per kilogram of body weight in a 15-year-old boy having 2.3 g of potassium per kilogram of body weight. His burden of Cs^{137} radioactivity was therefore approximately 4 percent of the K^{40} radioactivity naturally present in his body. Calibration of K^{40} and Cs^{137} body-burden measurements were made by using sacks of sugar mixed with weighted amounts of potassium chloride crystals, and a plastic dummy filled with water containing a known amount of dissolved Cs^{137}. In these determinations corrections were made for the contribution of scattered 1.46 Mev K^{40} photons to the 0.60 to 0.72 Mev band for 0.66 Mev Cs^{137} photons. However, no such corrections could be made for the contributions from other nuclides since we lacked data on the spectra of these isotopes when they were distributed in dummies of sizes similar to the children's bodies. Thus the burdens of combined Zr^{95} and Nb^{95} and of $Ru^{103, 106}$ could not be measured with any accuracy. However, they were less than the Cs^{137} burdens except in three cases where external contamination was suspected.

The thyroid glands of eight of these children were examined for I^{131} by placing two balanced detectors (12.5 by 10 cm) 5 cm in front of and behind the neck and counting for 30 minutes. Except for K^{40}, no well-defined peaks were observed. Therefore, I^{131} contamination, if any, was less than 200 pc on the basis of calibration with a set of identical hollow dummies of the thyroid gland filled with various standard concentrations of I^{131}. In these dummies a photopeak in the 0.30- to 0.40-Mev band was still visible at 200 pc and the increase of counting rate in the band was more than three times the standard deviation of the difference between the mean counting rates with and without the I^{131} source down to 50 pc.

Twenty-eight newborn infants, 6 to 24 hours of age, were examined during the period 10 May to 6 December 1962. The size and shape of the detection equipment were chosen by using the ratio (sample counting rate)^2/(background counting rate) as a figure of merit; as a result the optimum arrangement in our facility was placement of a single (12.5 × 10 cm) unshielded NaI detector 1.3 cm directly below the transparent plastic bassinet in which the infant was placed in a prone position for a maximum counting time of 45 minutes. The lower limit of detectability for I^{131} was 50 pc. An attendant always sat in the steel cubicle about 1.2 m away from the bassinet, and appropriate aseptic precautions were observed for the infants' protection. A second count was always taken immediately afterward, during which the attendant, empty bassinet, and cover sheet were counted for an equal time in the same relative

positions, as "background" to be subtracted from the gross counts obtained when the infant was present. The only radionuclide detected during the 45-minute counting period was natural K^{40}. This observation was especially interesting inasmuch as all of more than 200 adults and children examined here have had easily demonstrable body burdens of Cs^{137}, as well as the ubiquitous K^{40}. This was also true for the mothers of five of the newborn babies examined (*3*). The other 23 mothers were not examined for various reasons.

It was not feasible to maintain newborns in the counting room for sufficient time to show the possible presence of very small quantities of gamma-emitting radionuclides. However, seven stillborns (third trimester) were counted for a period of at least 10 hours each, with the detector equipment just described. Appropriate background and calibration measurements were taken (*4*). Excellent full-energy peaks for K^{40} were obtained from all seven stillborns. For example, based on a K^{40} calibration by means of a plastic dummy containing sugar and 12 g of potassium as KCl crystals mixed in, the potassium content of one infant weighing 3.23 kg was estimated to be 5.1 g. This was in close agreement with a value of 5.2 g predicted from the data of Anderson and Langham (*5*). No other radionuclides were found in three of the stillborns but in the others well-defined peaks corresponding to combined Zr^{95} and Nb^{95} and $Ru^{103, 106}$ were observed.

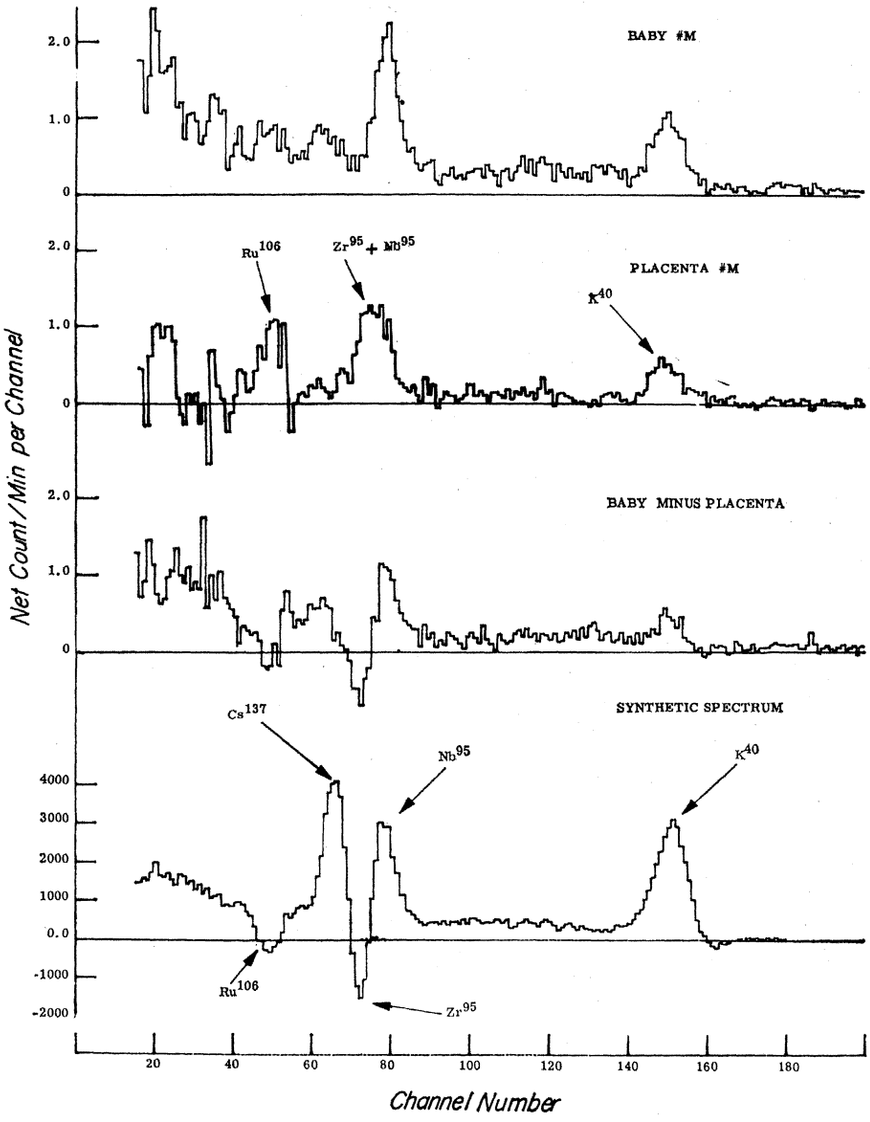

Fig. 1. Gamma spectra after background corrections. Counting times were 10 hours each, except for the synthetic spectrum, which was obtained by placing a dummy containing pure sucrose over the crystal detector and counting 2 kg of KCl plus authentic sources of Ru^{106}, Cs^{137}, and Nb^{95}, and subtracting background, thus simulating the net spectrum for a baby. The placenta spectrum was then simulated in the same way with less KCl and Ru^{106}, no Cs^{137}, and Zr^{95} in place of Nb^{95}. When this spectrum was subtracted, channel by channel, from the simulated baby spectrum the "synthetic spectrum" resulted. The shape of the real "baby minus placenta" spectrum differs mainly in having very little Cs^{137}. The low counting rates in the top three diagrams are significant because the counts were made over a 10-hour period.

The placental tissues were also obtained for three of these cases and were each counted for 10 hours in the same geometry. The histograms in Fig. 1 are typical of all three cases. When the net count/min per channel in the whole placenta was subtracted from the net count/min per channel in the whole baby, negative values were obtained in the locations corresponding to $Ru^{103, 106}$ gammas and Zr^{95} gammas. Thus each of these babies contained less of these isotopes than its placenta. However, they contained more Nb^{95} and K^{40} than their placentas. Since Nb^{95} is the radioactive daughter product of Zr^{95} disintegration and the ratio of Zr^{95} to Nb^{95} differed in baby and placenta, even on a basis of activity per gram of wet tissue, we must conclude that the human placenta discriminates between these two atomic species during their transport into the fetus.

The gamma emission spectra of Zr^{95} and Nb^{95} are so similar that they cannot be resolved in a mixture of the two nuclides. However, after physical separation by means of an anion-exchange column (*6*) the pure spectrum of each nuclide was obtained. When the spectrum of Zr^{95} was subtracted from that of Nb^{95}, the pattern shown in the bottom histogram of Fig. 1 resulted. The similarity of the Zr^{95}–Nb^{95} region to that in the "baby minus placenta" spectrum is striking. Cesium-137 was also put into the synthetic spectrum to indicate the normal location of this isotope's total absorption peak (0.66 Mev). The ratio of Cs^{137} to K^{40} in the stillborn babies was lower than for children and adults. This observation could be explained by postulating a placental discrimination against Cs when compared with K. Further data are being collected to verify these relationships.

A special effort was made to detect radioiodine in thyroid glands removed at autopsy during the period April through September 1962. Eighteen glands from fetuses ranging from 4 months to full term and six glands from infants from 1 day to 4 months of

age were examined. Each gland was counted in a 5- by 5-cm NaI, well-crystal detector with multichannel pulse analysis for 30 minutes. The minimum amount of I^{131} giving a detectable photopeak with this equipment was 30 pc. No gamma radioactivity of any sort was detected in any of these glands. That is, in the energy range from 0.06 to 2.0 Mev, the net counting rate was not significantly above background and no peaks were present. The natural K^{40} in the glands was not observed because the amounts of tissue were too small.

During this April to September period, the milk supplies of several cities in the United States contained readily measurable amounts of I^{131} (*7*). However, several milk samples from the Los Angeles area were examined by us and found to contain no I^{131}, a result not unexpected inasmuch as there was no local rainfull during that period. Furthermore, the dairy stock in the area is fed stored hay and grains, rather than open pasturage, a procedure which permits time for decay of the relatively short-lived radioiodines (*8*).

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2. From the Laboratory of Nuclear Medicine and Radiation Biology of the Department of Biophysics-Nuclear Medicine; and from the Department of Radiology, Obstetrics-Gynecology, and Pathology, School of Medicine, University of California at Los Angeles; and the Los Angeles County Harbor General Hospital, Torrance, California. Supported by contract AT(04-1)-GEN-12 between the U.S. Atomic Energy Commission and the University of California at Los Angeles.
3. Cesium-137 concentrations in 155 Los Angeles residents ranged from 9 to 160 pc per gram of body potassium, with a mean of 43 pc.
4. The sensitivity was 2.7×10^3 count/min per microcurie of K^{40} in a band 200-kev wide in which the background was 24 count/min. For Cs^{137} the values were 2.2×10^4 count/min per μc, background 104 count/min in a 200-kev band, and for I^{131}, 5.8×10^5 count/min per μc in a 110-kev band with a background of 90 count/min.
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6. L. Wish, *U.S. Naval Radiol. Defense Lab. Report TR-185* (1957).
7. U.S. Public Health Service Official News Release HEW-W21, 12 January 1963.
8. We thank M. Helper, E. Flynn, P. Suzuki, R. Hamel, E. James, O. Reiner, and R. Van Dusen for technical assistance.

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Mössbauer Spectrum of Iron-57 in Iron Metal at Very High Pressures

Abstract. *The effect of pressure on the Mössbauer spectrum of Fe^{57} in iron metal has been studied as the pressure was increased presumably to more than 140 kbar. At pressures up to 120 kbar, a six-line spectrum characteristic of α-iron was observed. At 140 kbar, a seventh line appeared in the spectrum at -0.12 ± 0.06 mm/sec relative to stainless steel. This line was attributed to the appearance of the high-pressure phase of iron.*

This paper reports on the development of a technique for conducting Mössbauer experiments (*1*) on materials under very high pressure and on the results obtained from a study of those changes produced by pressure in the Mössbauer spectrum of Fe^{57} in metallic iron. The effect of pressure on the Fe^{57} Mössbauer spectrum in iron metal is of interest for several reasons. All the nuclear parameters upon which the Fe^{57} spectrum depends can be determined from other experiments. Thus, the spectrum can be used as a probe into non-nuclear effects to which the spectrum is sensitive, such as the electronic charge density and spin polarization at the nucleus. The Mössbauer spectrum of Fe^{57} can be used both to detect and to characterize the reported high-pressure phase of iron metal (*2*, *3*). Furthermore, the dependence of the spectrum on the magnetic field at the nucleus can be used to study the effect of compression on the ferromagnetism of α-iron. Few other high-pressure experiments can yield such detailed information about behavior on an atomic scale, which makes the Mössbauer effect a most useful experimental tool for high-pressure research.

The theory of the Mössbauer effect and conditions for observation of recoil-free radiation have been worked out quite thoroughly (*4*) and will not be discussed here. Two experiments observing Mössbauer spectra in solids at elevated pressures have been reported. In a precise study (*5*) of the shift in the photon energy (the chemical shift) produced by hydrostatic pressures up to 3 kbar, most of the shift was attributed to the increase of the electron density at the nucleus. Other effects of pressure on the fine structure of the spectrum were not reported. The influence of pressure on the Mössbauer effect of Dy^{161} (dysprosium) formed *in situ* in Gd (gadolinium) metal has also been studied. The theoretical prediction made by Hanks (*7*) that higher pressure would permit observation of recoil-free radiation not detectable at one atmosphere was demonstrated, and very dramatic changes of the fine structure were found.

Since the 14-kev radiation would be attenuated severely in passing through the walls of the pressure vessel, it was decided to contain the radiation source under pressure in order that the radiation pass through the wall of the pressure vessel only once. For simplicity, the source was a disk of iron metal 0.175 inch (0.445 cm) in diameter and 0.007 inch (0.017 cm) thick. In order to minimize the absorption of the recoil-free radiation within the disk, it was made of iron enriched to 99.9 percent in Fe^{56}.

The 14-kev excited state of Fe^{57} was obtained from the decay of Co^{57} by electron-capture. The long half-life (270 days) of this decay controls the intensity of radiation which is essentially constant for a period of a few hours. To obtain the Fe^{57} in an iron rather than a cobalt environment, the Co^{57} was electroplated onto the iron disk; and the sample was annealed at 1000°C for 2 hours in an atmosphere of hydrogen after electroplating. A sample (*8*) with 4 mc of Co^{57} plated on the 0.007-inch (0.017-cm) edge along one-half of the circumference was used for the measurements reported here. Assuming a uniform distribution of Co^{57} within 0.005 inch (0.013 cm) from the edge, the cobalt concentration should be less than 0.1 percent.

The iron disk was contained between a set of Bridgman anvils, with face diameter of 0.250 inch (0.63 cm), by a ferric oxide coated ring of pyrophyllite 0.031 inch (0.079 cm) thick and 0.010 inch (0.025 cm) high. The Fe_2O_3 with which the ring was coated was enriched to contain 99.9-percent Fe^{56} to minimize absorption of the 14-kev radiation. Details of the ring and anvils have been described elsewhere (*9*). Pressure was applied with a 200-ton capacity hydraulic press through a calibrated, strain-gauge instrumented, load cell. This permitted continuous monitoring of the load on the anvil faces, which was constant within at most 2 percent at any pressure.

No information is available about the distribution of pressure in an iron disk in the Bridgman anvils, so pres-