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THE USE OF URANIUM IN CERAMIC TABLEWARE

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Uranium was first identified in 1789 by the German chemist Klaproth. It was used shortly afterwards in coloring of glazes and glass. As early as 1896, as the result of fogged photographic plates, it was ascertained that uranium was radioactive.

Lenchner (1926) discussed the use of various uranium compounds as an additive in the manufacture of greenish or yellow glass and glazes. This was further developed by Lorah (1927) who refined the technique. Shortly thereafter, McMaster (1932) obtained a patent for various formulas containing uranium to produce different hues in glazes.

The U.S. Atomic Energy Commission (1961) permitted the use of "exempt" quantities of uranium not to exceed 20 percent in the glaze of ceramic tableware and 10 percent in glassware. It was the opinion of the AEC that these levels of uranium did not constitute a significant radiation hazard. However, according to Crawley (1959), uranium-containing dishes were being used as "convenient sources" in high school radiation measurement experiments in the late 1950's. Public alarm escalated in the early 1960's as reflected in several news articles with such headlines as "Radiation Danger in the China Closet," "Radioactive Dishes Buried - in Backyard," "Hot Pottery Breakup," etc.

In 1963, the New York regional office of the Public Health Service obtained some radiation measurements of a typical place setting of uranium-containing dishes. Survey-meter open-window readings ranging from 15 mR/h at contact to 3 mR/h at 1 inch were reported. A person seated before such a setting would receive ~3 mR/h beta and gamma exposures to the hands and arms. The exposure to the torso at 25 cm would be less than 0.1 of this value.

Similar readings were recorded from a sample of Fiestaware collected in December 1976 by the Radiological Health Representative of the Philadelphia district office of the Food and Drug Administration (FDA). Using a GM survey meter, open-window readings at contact ranged from 5 to 7 mR/h with the highest readings obtained from the inside areas of a soup bowl. The reading at ~25 cm from a dinner plate (23.5-cm diameter) was ~0.7 mR/h.

A plate analyzed by the Bureau of Radiological Health, FDA, indicated the Ge(Li) spectrum of Figure 1, which confirmed the presence of natural uranium in the glaze.

An estimate of the skin dose due to beta emissions from the surface glaze may be made following a few conservative assumptions using the formula for beta dose from a point source to the arms and hands of an individual exposed to the beta radiations. For a single beta emitter,

$$D(\text{rad}) = \frac{4.1(t)(b)(C)(P\beta)}{x^2}$$

235 U

234 Th

10⁶ L

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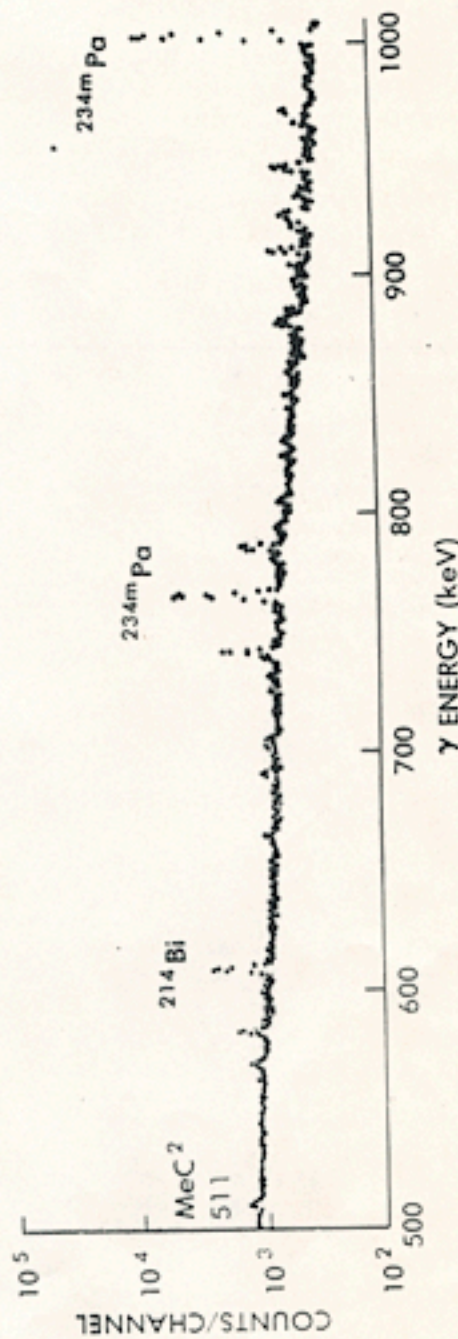
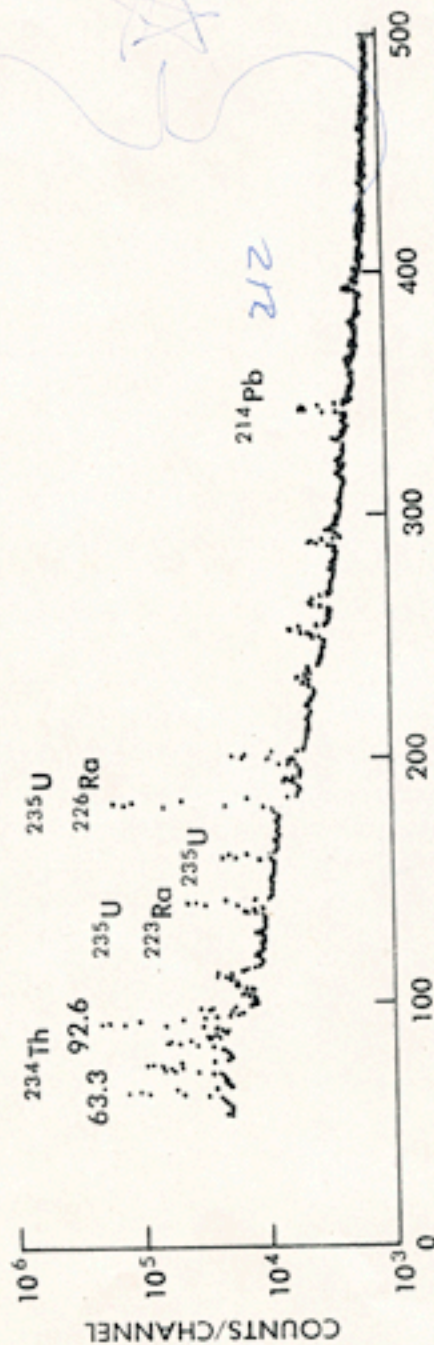
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where

4.1 is a constant arising from the choice of units for the variables in the equation
 t is the exposure time in days and is taken as 45.6, which represents 3 hours per day per year,

b is the fraction of disintegrations that result in beta emission,

C is the activity of the source in curies,

X is the distance from the source to the arms and hands which is taken to be 7.6 cm (3 in), and

P_β is the mass stopping power, given by

$$P_\beta = \frac{(10^6)(\mu_m)(\bar{E})e^{-\mu_m X}}{\rho_m}$$

where

μ_m is the absorption coefficient in cm^{-1} for betas in tissue,

\bar{E} is the average energy in MeV of the beta emitter,

x is the depth of tissue in cm, in this case equal to 0 since we are interested only in the surface of the skin, and

ρ_m is the density in g/cm^2 of the skin.

In computing the dose, beta emitters with maximum beta energy less than 70 keV were eliminated from consideration since these betas would be completely absorbed in the 3 inches of intervening air. The doses from the five remaining beta-emitting daughters are indicated in Table 1. Assuming them to be present in equal activities the total dose rate is ~ 2.4 rem/yr, which is $1/3$ of the 7.5 rem/yr maximum permissible dose (MPD) to the arms and hands of the general public as given in 10 CFR Part 20.

TABLE 1

RADIONUCLIDES IN TABLEWARE
BETA ENERGY DOSES

U-DAUGHTER	β -MeV	rad/yr
^{234}Th (UX_1)	0.19	0.97
^{234}Pa (UX_2)	2.31	0.27
^{214}Pb (Ra-B)	0.70	0.49
^{214}Bi (Ra-C)	0.60	0.25 (77%)
^{210}Bi (Ra-E)	1.17	0.38 (23%)
		<hr/> 2.42

O'Donnel et al. (1974) estimated the total dose to the population assumed to be possible due to the distribution, use, and disposal of tableware containing radioactive material if used in eating establishments. Included in the study was tableware assumed to contain 20% natural uranium in the glaze. It was also assumed that such radioactive tableware was used in restaurants instead of the usual nonradioactive dishes. In addition, assumptions were made as to the duration of direct external exposure times of various subjects involved in the use of such dishes. Of these it was calculated that dishwashers, exposed for 2000 hours, would receive 34.4 mrem/yr; waiters, at 1000 hours of exposure, would receive ~7.9 mrem/yr; and a patron seated before a setting of radioactive tableware for a total of 4 hours would receive no more than 0.2 mrem/yr.

Claus (1958) suggests uranium as "a powerful chemical nephrotoxin." Once absorbed in the bloodstream, uranium, as the hexavalent ion, complexes with protein and bicarbonate. The diffusible bicarbonate complex enters the glomerular filtrate. In the proximal tubules, the complex breaks down freeing the uranyl ion which, by blocking carbohydrate metabolism, injures the epithelial cells. According to Claus, the injury is a chemical one; it is independent of the amount of radioactivity, and the effect is acute. Fortunately, only trace amounts of uranium are absorbed from the GI tract and the skin. The International Commission on Radiation Protection (1964) made special mention of the chemical toxicity of natural uranium saying "... the ingestion (of soluble uranium) averaged over two days should not exceed 150 mg..."

Converting the ICRP maximum permissible concentration for uranium in water (MPC)w of 6×10^{-7} $\mu\text{Ci/ml}$ chronic intake for the general public to mass units, a value of 1.8 mg/l is obtained. Over a 13-week period at 2.2 l/day this equates to 360 mg intake. The National Committee on Radiation Protection and Measurements (1963) cites an (MPC)w of 7×10^{-4} $\mu\text{Ci/ml}$ based only on radiotoxicity. The more conservative ICRP value will be followed in this paper.

In 1971 an inspector from the Detroit district of the FDA collected some samples of red pottery dishes. In addition to radiation measurements, analysts in the Bureau of Foods, FDA, conducted 24- to 60-hour "soak" experiments using 4 percent acetic acid solutions to duplicate storage of certain acid foods such as sauerkraut. A concentration of 55 ppm of uranium was reported to be present in the 50-hour leach solution. This was equivalent to 1.66×10^{-5} $\mu\text{Ci/ml}$ for uranium. This exceeds the (MPC)w of 6×10^{-6} $\mu\text{Ci/ml}$ occupational exposure recommended by the ICRP by a factor of 2.7, or 27 for the public, from the continuous ingestion of drinking water based on chemical toxicity of uranium. However, according to the Bureau of Radiological Health (BRH) investigators, following the assumed aforementioned short-term limit recommended by the ICRP, the ingestion of 100 ml of the 60-hour leach solution averaged over a 2-day period would constitute an intake of ~7% of the permissible 150 mg.

The International Expert Committee on Food Additives (1972) published some permissible chronic intake levels for cadmium, mercury, and lead (Table 2). The ICRP (1964) limit for uranium is included for comparison. As indicated, cadmium is the most toxic of the four, mercury next, with lead and uranium being the least toxic of these elements. However, it was the opinion of the BRH that the exposure levels of uranium, albeit admittedly low, constituted an unnecessary and avoidable exposure to the public. In addition, based on the leach data, such tableware is subject to the food additives clause of the Food, Drug, and Cosmetic Act and thus subject to regulations.

max. ceiling
170 mrem/yr
U.S.

34.4 = 1/5 allowable

+ 100 mrem/yr
Background
avg.

134.4

rad/yr

0.97

0.27

0.49

0.25 (77%)

0.38 (23%)

2.42