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NUCLEAR ACTIVATION WITH COBALT-60 GAMMA RAYS

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NUCLEAR ACTIVATION WITH COBALT-60 GAMMA RAYS

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ABSTRACT

Twenty-four elements that occur in foods have been tested for induced activity after irradiation in a multi-kilocurie source of cobalt-60. No activity detectable with a scintillation well counter was observed.

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INTRODUCTION

It has been assumed by many who have studied the effects of x-rays and gamma rays on food sterilization, chemical reactions, etc. that these radiations induce no radioactivity. This assumption is based on the fact that the irradiated materials show no activity when tested with ordinary radiation monitors. In most cases, however, the background radiation level is such that small amounts of induced activity could be easily overlooked.

A search of the literature indicates that x-rays can indeed induce low levels of activity in certain elements. A 4.1-hour indium activity was reported (1) first in 1939 by x-ray excitation, and was shown (2) to have a threshold for production of 1.2 ± 0.1 Mev. Later it was found that lead (3) and silver (4) activities could also be produced by x-rays with energies up to 1.5 Mev. Wiedenbeck (5,6,7) studied the excitation of these elements as well as other elements such as rhodium, cadmium, gold, and krypton. In each of these cases the ground state isotope was excited to an isomeric state by the x-radiation. The isomer then decayed by its characteristic half life and energy (8) as shown in Table I.

Guth (9) has shown that this excitation by x-rays is a "line absorption", in which x-rays of only a certain limited energy will induce the activity. It was found that the widths of the lowest nuclear levels of lead and indium are of the order of a few millivolts. Hence in the continuous spectrum of 1.5 Mev. x-rays only about 1 in 10^8 x-ray quanta has an energy suitable to induce activity.

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A logical extension of this work was to determine whether indium activities could be produced by the gamma rays from radium and its daughters. Guth (9) reports that Goldhaber, Hill and Szilard irradiated indium with the gamma rays from 1/2 gram of radium without producing any activity. Since it is known that the energy of these gamma rays is sufficient to excite the indium, the absence of activity must be attributed to the fact that none of its known gamma ray energies are equal to the energy of the activation level. The probability of excitation is therefore small and Guth concludes that only irradiation by a continuous x-ray spectrum is likely to lead to a line absorption.

With the advent of multi-kilocurie sources of cobalt-60 and eventual megacurie sources of fission products, gamma ray intensities many orders of magnitude greater than that from 1/2 gram of radium are available. Furthermore, the thickness of these sources tends to "smear" out the energy of the gamma rays emitted by the source until the gamma energy spectrum no longer shows sharp, nearly monoenergetic rays but instead a gamma ray distribution which approaches a continuous spectrum. This spectrum should then be nearly as effective as the continuous x-ray spectrum for activation. Indeed Harbottle (10) has recently published in this journal a report of the activation of indium and cadmium isomers with cobalt-60 gamma rays.

This paper is the initial report of an independent experimental program conceived as the result of discussions at the Symposium on Radiation Sterilization of Foods sponsored by the Committee on Foods of the Advisory Board on Quartermaster Research and Development. This symposium was held on June 26, 1953 at Massachusetts Institute of Technology under the general auspices of the National Research Council. This program is not designed to evaluate the tolerance limits of various activities in foods. Its purpose is only to determine within the limits

of detection available to it, the amount of activity (if any) induced in a number of elements by gamma rays from cobalt-60 and later from gross fission products. With such extensive effort being expended by a number of laboratories in trying to perfect the technique of food sterilization with gamma radiation, it was felt that some numerical limits for the amounts of activity induced by gamma radiation should be experimentally determined. Once determined, these values could then be evaluated as to their hazard by the proper authorities.

EQUIPMENT

A. The Cobalt-60 Source

The Fission Products Laboratory and the Michigan Memorial-Phoenix Project of the University of Michigan have been conducting experiments (11-15) for several years on the effects of gamma radiation on food sterilization and chemical reactions. A "1-kilocurie" cylindrical cobalt-60 source (16,17) obtained from Brookhaven National Laboratory, was used in early experiments. In January of 1953 a "10-kilocurie" cobalt-60 source (18,19) was obtained from the Chalk River Reactor. This source consists of 100 cobalt rods, 1/4" in diameter and 10 inches long, encased in aluminum pipe. The source is housed in a radiation room with 4 foot thick, solid concrete walls. The cobalt rods are mounted in a double circle in an aluminum frame and are lowered into a 16 foot well of water to "shut off" the radiation. When material is to be irradiated it is set-up in the radiation room and the source is raised into the room. A phantom view of the radiation chamber is given in Fig. 1. This "10-kilocurie" source gives a radiation flux of about 220,000 rep per hour over the 6 1/2" diameter area in its center. This flux corresponds to about 5×10^{10} photons per cm^2 per sec.

B. Counting Equipment

From the literature references it was apparent that if any activity

were formed it would be low level and would require a sensitive detector. In addition, Table I shows that the isomeric states formed by the activation decay, in general, by the emission of gamma rays. An ordinary geiger tube, whose counting efficiency for gamma rays is only about 1%, is ineffective in detecting such radiation. A scintillation well counter had been obtained however from the Nuclear Instrument and Chemical Company for other low level gamma ray counting work on a nuclear chemistry grant from the Atomic Energy Commission. This counter, made available for the gamma activation work, enabled us to increase the sensitivity of the measurements.

The well counter consists of a 1- 3/4" diameter, 2" thick crystal of thallium activated sodium iodide. A 3/4" hole is drilled 1 1/2" deep in the center of this crystal and the entire crystal is encased in 1/32" aluminum. A test tube containing a sample is inserted in the hole for measurement. Rays (beta and gamma) which penetrate the wall of the tube and the thin aluminum casing of the crystal are absorbed in the crystal, giving a flash of light. This scintillation is then "seen" by a Dumont 6292 (K-1186) photomultiplier tube which feeds a scaler and register. The particular well counter used had a counting efficiency of about 43% for cobalt-60 rays and about 51% for those of iodine-131.

PROCEDURE

Since meat is one product for which radiation sterilization may be particularly well suited, the trace elements it contains were checked for possible activation by the cobalt-60 gamma ray sources. The amount of various trace elements which occur in meat (beef) has been given in American Scientist (20) based on a method developed by Arthur J. Mittle-dorf of the Armour Foundation. He estimates that during a year the average American eats about 60 pounds of beef. The amount in ounces of trace elements a person obtains in eating these 60 pounds of beef

is given in Table II.

Pure samples of each of these elements (as well as carbon, oxygen, sulfur and iodine) were placed in test tubes 1.6 cm in diameter and 15 cm long which just fit into the counting well. The test tubes were filled with at least 2" of sample so that the well would be completely filled with the sample for maximum detection sensitivity. These samples were irradiated by the 10-kilocurie cobalt-60 source for varying periods of time and then counted in the scintillation well counter. Because of a large detection area this counter has a high background counting rate. This background was reduced to about 300 counts per minute however by enclosing the counter in about 4" of lead. Counts of one hour duration both for background and sample gave reasonable statistical precision. A count of this length gives a "9/10's error" of about 1.2% for the determination.

RESULTS

A. Indium Activation

The method of activation by gamma radiation was first tested by the irradiation of indium in the cobalt-60 source. It was felt that if the source had enough intensity to produce activity in anything it would activate the indium. The tube was filled with about 85 grams of indium and irradiated in a flux position of about 220,000 rep/hr, nearly to saturation of the 4.5-hour activity. When counted, the sample showed activity about forty times the background of the well counter. The activity decayed with a half life of 4.41 hours with a 9/10's reliability factor of 0.05 hours. This value agrees with accepted literature values for $\text{In}^{115\text{m}}$. A rough attempt to evaluate the efficiency of the well counter gave a value of about 50% for the $\text{In}^{115\text{m}}$ gamma ray. In addition a rough evaluation of self absorption of the radiations in the indium metal indicates that about 1000. γ dis/min/gm of indium were formed in the

irradiation or roughly 4.5×10^8 dis/min/gm In. per 1000 rep/hr saturation flux. Harbottle (10) has recently reported similar results in an independent investigation at Brookhaven National Laboratory. He reports an order of magnitude value for the cross section for formation of indium from cobalt-60 gamma rays of 10^{-9} barns.

B. Trace Element Activation

The work with indium was admittedly weighted heavily in favor of success but it established that the method was satisfactory to detect small amounts of activation induced by gamma radiation. Twenty-four samples of pure elements or compounds were then prepared and irradiated. The elements, their sources and weights are listed in Table III.

These elements were subjected to two series of experiments. In one the samples were irradiated simultaneously for a day or so and then rapidly removed from the source room and counted. It was possible by rapid manipulation of the source to have a sample in the well counter between 40-45 seconds after the end of irradiation. By raising the source immediately after removal of one sample, it was assured that any short lived activity in the remaining samples would remain at saturation. Short lived activities with half lives down to the order of 20 seconds could have been detected if formed in even one-tenth the yield of the indium. No activity greater than 5% of background however was found in any of these samples in twenty minute counts.

In a second set of experiments the samples were irradiated in the high flux position of the source at a flux level of 220,000 rep per hour for about two weeks. The samples were then removed individually and counted. They were compared with a background count of the same day and also with a blank sample count before irradiation. An hour background count fluctuated $\pm 1\%$ over the three week period of the measurements.

It was found that only two samples varied from this background region by more than $\pm 1\%$. One was potassium chloride whose natural radioactivity increased the blank count to two-thirds more than background. The irradiated sample gave the same count as the blank sample, however. The second sample was the chromium metal powder. An increase was found in this sample after irradiation but the activity did not decay over a period of several weeks and must be attributed to contamination.

DISCUSSION

It should be stressed that in the method used, the radiation must pass through the test tube, about 300 mg/cm^2 thick, and the aluminum casing of the crystal before it is detected. This then prevents the detection of gamma rays of less than about 30 Kev energy or of most conversion electrons from highly converted gamma rays. Since early work in this field pointed out several elements whose isotopes upon activation decayed with such weak radiations, the present work is somewhat limited. Nevertheless, it was thought worthwhile to perform the experiment with the equipment on hand and to recognize the fact that nuclides emitting certain very weak radiations would escape detection.

Activity found in a sample might also be accounted for by conditions other than gamma ray activation. One of the elements tested, potassium, has a low abundance isotope that is naturally radioactive and was detectable in the scintillation crystal. Natural radium also often occurs to a small extent in many materials. Shandley (21) at the University of Rochester has reported a highly sensitive method for determining the radium content of common foods by deemanation. He found that most of the foods tested fell within the range of 0.74 to 6.5×10^{-15} gram radium per gram of starting material. This amounts to about 10^{-9} microcurie of radium or about 2×10^{-3} disintegrations per

minute per gram of food. The use of high purity samples of elements in our experiments reduced the possibility of such contamination. In addition as a check on the above items, careful background blank counts were taken on each sample and compared with the activity of the sample after irradiation.

SUMMARY

This work substantiates the belief that no radioactivity is formed when 24 elements common as trace elements or substituents in food are irradiated in a "10-kilocurie" cobalt-60 source. Two extensions of the work, however, should be made. For one a sensitive method should be utilized for detecting the weakest radiations without self absorption and container absorption effects. The use of a relatively large volume of a liquid scintillator such as used in low level carbon-14 and tritium tracer work (22-24) might give the required detection sensitivity in this weak radiation range. Chemical problems of dissolving the element and scintillator in a mutual solvent would, however, add difficulties to this approach.

In addition, this work was further limited by the fact that it was done with cobalt-60, whose two gamma rays have an energy of 1.33 and 1.13 Mev. Any commercial method of food sterilization would probably use gross fission products in some form. The gamma spectrum emitted by such a source depends upon the age of the fission products and the energy of the neutrons producing the fission. The contribution of radiations from particular fission product isotopes has been presented as a function of age for U^{235} thermal neutron fission by Hunter and Ballou (25). From their work it can be assumed that many of the gamma rays will have energies equal to or lower than those of the cobalt-60 radiations.

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However small percentages of higher energy gamma rays may be present in the fission products from an operating reactor after a few weeks or months of "cooling". The energy spectrum of "thick" sources of these products thus could start at a much higher energy than the cobalt spectrum. Since the early x-ray work showed that a number of elements were capable of excitation at energies between 1.5 and 3.0 Mev. the experiments reported here should be repeated with kilocurie or larger sources of fission products. Suitable sources now exist in the gamma ray facility of the Materials Testing Reactor, where spent fuel rods from the reactor are allowed to "cool down" before processing. Samples can be irradiated here in an area of high flux fission product radiation.

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TABLE I⁽⁸⁾

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Nuclear Data for Isomers

Isomer	Half life	Radiation	Energy of Photons (Mev)	Gamma rays Converted
Rh 103 m	57 min.	γ	0.040	highly
Ag 107 m	44.3 sec	γ	0.094	$e^-/\gamma \sim 16$
Cd 111 m ₂	48.6 min.	γ	0.150	$e^-/\gamma = 2.3$
			0.246	$e^-/\gamma = 0.064$
In 115 m	4.50 hr.	γ (95%)	0.335	$e^-/\gamma = 0.98$
		β^- (5%)	0.83	
Au 197 m	7.4 sec.	γ	0.130	$e^-/\gamma \leq 2.0$
				K/L/M = 1/7.5/3.6
Pb 204 m ₂	68 min	γ	0.279	$e^-/\gamma \sim 0.3$
			0.905	$e^-/\gamma \sim 0.1$
			0.374	$e^-/\gamma \sim 0.05$

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OUNCES OF TRACE ELEMENTS IN ONE YEAR'S CONSUMPTION (60 lbs) OF BEEF

Aluminum	0.00001	Molybdenum	0.000014
Boron	0.00006	Nickel	0.000003
Calcium	0.0025	Phosphorus	1.6
Chromium	0.000003	Potassium	4.7
Cobalt	0.000002	Silicon	0.00006
Copper	0.00043	Silver	0.000013
Iron	0.0306	Sodium	0.40
Lead	0.00001	Tin	0.000002
Manganese	0.00002	Zinc	0.045
Magnesium	0.204		

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TABLE III

FORM, SOURCE, AND WEIGHT* OF ELEMENTS IRRADIATED IN GAMMA

RAY SOURCE

1. Aluminum powder - High purity - Alcoa. - 2.7 grams
2. Elemental Boron powder - Student prep. - small amount of chemical contamination - 4.8 grams.
3. Calcium metal turning - B and A reagent grade - 2.1 grams
4. Carbon - graphite flakes - Fisher - 2.9 grams
5. Chromium metal powder - Charles Hardy - 15.1 grams
6. Cobalt metal powder - Old Kahlbaum material - contains some Cl - 5.9 grams
7. Copper metal pellets - Standard Laboratory grade - 16.3 grams
8. Iron metal powder - Standard Laboratory grade - 12.2 grams
9. Elemental iodine crystals - Mallinckrodt analytical reagent - 11.0 grams.
10. Lead metal filings - Mallinckrodt analytical reagent - 21.5 grams
11. Magnesium metal powder - Dow - high purity - 5.5 grams
12. Manganese metal powder - Charles Hardy - 15.1 grams
13. Molybdenum metal powder - Primus Chem. Co. (1912) - 14.5 grams
14. Nickel metal shot - Mallinckrodt analytical reagent - 16.1 grams
15. Oxygen - (Al_2O_3) - Harshaw - 5.3 grams
16. Phosphorus - $(NH_4)_2HPO_4$ - Mallinckrodt analytical reagent - 3.7 grams
17. Potassium - KCl - General Chemical Co. - reagent grade - 5.3 grams
18. Elemental silicon powder - Fairmount Chemical Co. - 4.0 grams.
19. Silver metal powder - G. Frederick Smith - reagent grade - 16.0 grams
20. Sodium and Chloride (NaCl) Merck analytical reagent - 5.9 grams
21. Sodium and Fluoride (NaF) - C.P. - 3.3 grams
22. Sulfur (roll) - cast - 4.38 grams
23. Tin metal granules - General Chemical Co. - reagent grade - 15.1 grams
24. Zinc metal granules - C.P. - 10.6 grams.

* The weight listed in the table is the amount of material contained in the bottom one centimeter of the test tube used in the irradiation and counting.

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Fig. 1
Phantom view
of radiation chamber for "10 kilocurie" cobalt-60 Source.

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