APR 24 1964

UCRL-7761

CONF-470-7

MASTER

Characteristics of radioac- tivity produced by nuclear explosives

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Facsimile Price \$ 1.60

Microfilm Price \$.80

Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

third plowshare symposium
engineering with
nuclear explosives

freeborn hall

university of california, DAVIS

april 21 - 22 - 23, 1964

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CHARACTERISTICS OF RADIOACTIVITY PRODUCED BY
NUCLEAR EXPLOSIVES

John A. Miskel

Lawrence Radiation Laboratory, University of California
Livermore, California

BIOGRAPHICAL SKETCH OF AUTHOR

B.S., Chemistry, University of California, Berkeley, June 1943.
Ph.D., Chemistry and Physics, Washington University, St. Louis,
Missouri, 1949 (Gladys Hermanns scholar; AEC Fellow).

1943-1946 Los Alamos Scientific Laboratory: Neutron excitation
functions, heavy element chemistry, fission process studies.

1949-1955 Brookhaven National Laboratory: High-energy nuclear
reactions, atomic effects of nuclear decay, nuclear decay schemes.

1955- Lawrence Radiation Laboratory, Livermore: Radio-
chemical diagnostics, nuclear spectroscopy, investigations of the
fission process, excitation functions, studies of debris from nuclear
devices.

1961 OEEC Sr. Visiting Research Fellow, Nobel Institute
of Physics.

ABSTRACT

The amounts and kinds of radioactivities produced by detonation of a nuclear explosive are dependent upon the specific design of the explosive. The two design extremes are a pure fission device, which will produce about 1.4×10^{23} fissions, and therefore about 2.8×10^{23} fission products, per kiloton of energy released, and a pure thermonuclear device which would produce no fission products, but would produce approximately 10^{23} atoms of tritium per kiloton. In both cases, interaction of escaping neutrons with the materials of the device itself, and with the surrounding media, could lead to further radioactivities.

The behavior and ultimate fate of the activities produced by the explosion depend on the composition of the medium in which the detonation occurs, the nature of the detonation, and the chemical species involved.

Some typical cases are described.

CHARACTERISTICS OF RADIOACTIVITY PRODUCED BY NUCLEAR EXPLOSIVES

John A. Miskel

The production of energy by nuclear reactions results in the production of radioactive nuclei. Therefore, in considering the possible utilization of nuclear explosives for peaceful purposes it is necessary to be able to predict the expected activities, their amounts, and dispositions.

These activities may arise from two sources: (1) They may be a direct result of the energy producing nuclear reaction, or (2) they may be the result of the interaction of a product of that nuclear reaction with some other material. The obvious example of the first source is the fission products which arise in the fission process; the activation of the environmental materials by neutron capture is an example of the second source.

The total radioactivity resulting from an explosion depends, of course, on the detailed design of the device and on the specific environment in which it is detonated. The device can range from one where energy is derived solely from fission to, ideally, one whose energy is derived solely from thermonuclear reactions.

In the fission process, two fragments are produced with a total kinetic energy of about 170 MeV. After losing their internal excitation energy, these fragments undergo on the average about 3 beta decays before reaching stability. The six beta decays per fission release an additional energy of about 21 MeV divided between gamma rays (about 5 MeV), beta particles (about 6 MeV), and neutrinos (about 10 MeV). In addition, about 2.5 neutrons with an average kinetic energy of 2 MeV are emitted per fission. If one considers only the prompt energy from fission, about 180 MeV per fission, there are 1.46×10^{23} fissions per kiloton.

A kiloton of fission therefore produces 2.9×10^{23} fission fragments; these fission products are distributed in mass according to the well-known fission yield curve shown in Fig. 1. Although the fission yield curves differ somewhat in detail for different fissioning nuclei, the general shape is the same for neutron-induced fission of U^{235} , U^{238} , and Pu^{239} . In the fission process the directly formed fragments are generally neutron rich. Therefore, they beta decay in order to become more stable. Since each beta decay process increases the nuclear charge by one unit, the chemical species changes with each decay. The physical-chemical behavior of the radioactivity resulting from fission is therefore strongly dependent on the time scale.

The excess neutrons produced in the fission process (about 1×10^{23} neutrons per kiloton) ultimately interact with matter, either the structural materials of the device or the materials of the immediate environment, and may result in the production of other radioactive species. In general these activities are produced by neutron capture (n, γ reactions) and decay directly to a stable isotope. Their chemical behavior is not, therefore, time dependent in so far as their radioactivities are concerned.

The fusion process, the energy source in thermonuclear devices, results in approximately ten times as many neutrons per kiloton as does the fission process. These neutrons undergo reactions similar to those of the neutrons resulting from the fission process although the higher initial energy of these neutrons increases the probability for ($n, 2n$) and (n, p) reactions. In addition to the neutrons, tritium is produced in relatively large amounts, on the order of 7×10^3 to 5×10^4 curies per kiloton of fusion.

Since the behavior of the fission products is dependent on their chemical nature, the percentage contribution of various elements to the total fission activity for several times is given in Table I. The various elements have been grouped into volatile, refractory, and intermediate.

In a cratering experiment the expected fate of the various activities can then be roughly predicted by consideration of the time at which the

Fig. 1. Yield-mass curve for thermal fission of U^{235} .

Table I. Percent contribution of elements to total activity produced in fission.

	Element	1 sec	10 sec	30 sec	1 min	5 min	10 min	1 h	1 day
Volatile	Kr	3.6	3.8	5.0	5.8	4.3	2.5	4.2	—
	Xe	4.6	2.6	3.8	4.6	9.0	5.0	3.5	14
	Br	2.5	3.3	5.0	6.0	2.2	—	1.0	—
	I	3.4	4.0	6.0	7.0	3.5	3.5	7.0	16
	Te	1.9	2.7	2.5	2.0	3.5	9.0	14	3.9
	Se	1.3	1.5	1.0	—	—	—	—	—
	Total	17.3	17.9	23.3	25.4	22.5	20.0	29.7	33.9
Intermediate	Sn	1.3	1.1	—	—	—	—	—	—
	Sb	—	1.0	2.3	3.7	5.0	2.3	2.0	—
	Cs	9.5	7.0	7.0	7.0	8.0	8.0	8.0	—
	Rb	7.0	8.0	8.0	8.0	11.0	7.0	4.8	—
	Mo	—	3.0	6.0	6.0	5.0	7.0	3.0	4.2
	Tc	—	1.2	4.0	8.0	5.0	9.0	7.0	—
	Total	17.8	21.3	27.3	32.7	34.0	33.3	24.8	4.2
Refractory	Ba	11	7.0	2.6	2.4	8.0	9.0	8.0	1.1
	Sr	11	6.0	4.0	5.0	11	4.0	3.7	6.5
	Y	15	11	9.0	8.0	4.6	7.0	5.0	13
	Zr	12	12	7.0	3.5	—	—	—	8.6
	La	7.0	9.0	8.0	4.0	2.8	9.0	10	1.6
	Nb	5.0	10	12	11	8.0	5.0	6.0	18
	Ce	2.5	4.7	3.0	1.0	2.5	5.0	4.4	6.6
	Pr	—	1.7	3.7	5.0	4.6	3.7	5.0	2.9
	Nd	—	—	—	—	—	—	1.0	—
	Total	63.5	61.4	49.3	39.9	41.5	42.7	43.1	58.3

GLL-643-744

debris is vented. The absolute amount of any species that emerges into the atmosphere will depend, of course, on the depth of burial and nature of the medium.

Some typical decay chains, which lead to isotopes of interest from a health and safety standpoint are:

(The percentages in parentheses are the cumulative fission yield to that point in the decay chain.)

Thus, in a detonation that ruptured the surface to produce a crater in a matter of a few seconds, one would expect the Sr^{89} and Sr^{90} to behave in the manner of their volatile precursors and be enriched in the vented debris. Iodine¹³¹, with its precursors, would behave in a manner more characteristic of the intermediate groups than of the volatiles.

The effect of the medium in which the detonation occurs on the fate of the activities is tied to the chemical stability of the compounds that might form. In the extremely high temperature existing during the explosive phase, one expects almost all species to exist as

unassociated ions. During the early cooling phases, if the debris is exposed to the atmosphere, the stable oxides probably form, or perhaps the nitrides. If the cooling occurs while the debris is still contained, the system may be in a reducing atmosphere leading to the formation of elements and hydrides rather than of oxides and nitrides. The subsequent radioactive decay, if it occurs after venting, may result again in an oxide, or, if the density of inert debris is sufficiently high and the chemical affinity large enough, reactions with the media may occur resulting in silicates, aluminates, etc., depending on what is available. Another possibility is that an atom, after being formed by the decay of its parent, will collide with some inert material and stick to the surface. The real behavior is therefore quite difficult to predict with any degree of precision with the available data.

The behavior of the activities resulting from neutron capture reactions can be predicted in somewhat the same fashion as the fission products, but no consideration of the decay chains is necessary in general.

If we consider a device designed for a Plowshare application with a total energy of 1 megaton and assume it to be 1% fission and 99% fusion, then we can make some estimates of the various activities produced, and can speculate as to their ultimate fates.

The fission involved would be 10 kilotons, with the associated 2.9×10^{24} fission products and about 10^{24} neutrons. The fusion would produce about 10^7 curies of tritium and 1.4×10^{27} neutrons.

As a first approximation, we can assume that the device is surrounded by a nonactivating neutron absorber, such as boron, which reduces the emergent neutron flux by a factor of 100. The total number of neutrons out is then about 1.4×10^{25} .

The activation products resulting from the neutrons depend on the medium; Plateau Basalt, the composition of which is given in Table II, has been used to calculate the activities to be expected from the detonation of a device such as we have assumed.

Table II. Composition of Plateau Basalt.

Oxide	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	FeO	MnO	MgO
wt %	48.8	2.19	13.98	3.59	9.78	0.17	6.70
Oxide	CaO	Na ₂ O	K ₂ O	H ₂ O	P ₂ O ₅		
wt %	9.38	2.59	0.69	1.80	0.33		

The fraction of the escaping neutrons reacting with any element in the surrounding medium can be calculated on the assumption that the neutrons are essentially thermalized before reacting and then using the known thermal neutron capture cross sections. In Table III are given the percentages of the neutrons captured by the various elements in

Table III. Percentage of neutrons captured by various elements in Basalt.

Element	Si	Ti	Al	Fe	Mn	M _g	Ca	Na	K	H	P
% neutron capture	12	14.8	2.8	43.4	2.5	9.7	6.7	4.1	1.3	2.8	0.08

basalt, calculated in this way and assuming that oxygen does not contribute. In a medium with low water content, such as basalt, it might be better to use a somewhat higher value than thermal for the average neutron energy, at the time of capture. However, the relative cross sections, and therefore the fractions of neutrons captured, do not change drastically in most cases.

In basalt, several of the principal constituents lead to stable or quite short-lived isotopes. Thus, titanium, which accounts for 14 to 15% of the neutrons, leads to a 6-minute activity; aluminum, which accounts for 6%, a 2.3-minute activity; and magnesium accounts for 1% and has a 10-minute activity.

The predominant activities, with half-lives greater than a few minutes, produced by neutron capture in basalt are listed in Table IV. The total activities are based on 1.4×10^{25} neutrons being captured in the medium.

Table IV. Neutron activation products in basalt.

Activation Products	Nuclide	Half-life	Radiations	Curies at t_0
	Si ³¹	2.62 h	β^- , 1.47 MeV γ , 1.26 MeV (~0.1%)	7×10^7
	Fe ⁵⁵	2.7 yr	γ 0.006 MeV	7.5×10^5
	Fe ⁵⁹	45 day	β^- , 0.27, 0.46 MeV γ , 1.1, 1.28 MeV	3.8×10^4
	Mn ⁵⁶	2.58 h	β^- , 2.8, 1.0, 0.6 MeV γ , 0.85, 1.81, 2.13 MeV	8.4×10^8
	Ca ⁴⁵	165 day	β^- , 0.255 MeV no γ	4.3×10^4
	Na ²⁴	15 h	β^- , 1.4 MeV γ , k, 37, 2.76 MeV	1.9×10^8
	K ⁴²	12.4 h	β^- , 3.55 MeV, 1.99 MeV γ 1.53 MeV (~20%)	2.8×10^6
	P ³²	14.3 day	β^- , 1.7 MeV no γ	1.8×10^5
Fission Products	Sr ⁹⁰	28 yr	β^- , 0.45 MeV no γ	1.8×10^3
	Mo ⁹⁹	66 h	β^- , 1.2, 0.41 γ , 0.140, 0.75	6.7×10^6
	I ¹³¹	8 day	β^- , 0.61 MeV γ , 0.36 MeV	1.28×10^6
	Nd ¹⁴⁷	11 day	β^- , 0.81 MeV γ , 0.09 MeV	7.5×10^5

For comparison, the numbers of curies of several fission products produced by this device are also indicated in Table IV.

In addition to the isotopes listed in Table III, several of the light elements may undergo (n, α) or (n, p) reactions to produce activities. This is particularly true for the thermonuclear neutrons; considerable Na^{24} may be produced by Al^{27} (n, α), some Na^{22} by Na^{23} (n, 2n), and some scandium isotopes from titanium. The steels and other construction materials may produce other species such as Co^{60} , Co^{57} , Co^{58} , and Mn^{54} .

The amounts of activities tabulated above are the totals produced by the device. In a cratering-type application, only fractions of the amounts produced would be vented into the atmosphere. The specific fractions will depend on the chemical behavior of the substance involved. Using the crude classifications of Table I, it would appear (for a venting time of a few seconds) that even if one assumed all of the "volatiles" and half of the "intermediates" were in the atmosphere, and remained there after the fall-back of the major part of the total mass vented occurred, this would be less than one-third the total fission product activity produced. It would be expected that, after radioactive decay, the less volatile nuclides formed would be scavenged by the particulate matter as it falls out.

The induced activities from basalt, except for the sodium, potassium, and phosphorus, are "refractory."

The distribution of activities has two immediate consequences. One is related to the re-entry problem where, in order to do further construction or to utilize the results of the explosion, it is necessary for people to return to the active area. The second is related to the problem of introducing the activities into the biosphere.

The first problem is concerned primarily with the level of γ radiation, whereas all types of radiations are of interest for the second problem. A considerable amount of experience exists on prediction of

down-wind deposition and of radiation fields from clouds of known composition. There is a more limited experience in determining cloud composition from partially contained detonations.

Studies are under way at Livermore to investigate the system more thoroughly. In any forthcoming Plowshare experiments, a large number of measurements will be made in order to document the distribution of a representative variety of radioactive species. In addition, laboratory experiments on thermodynamic properties of the various elements are being conducted to enable better predictions of the high-temperature reactions and condensations phenomena to be made.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.