RADIOACTIVITY SOURCE TERMS FOR UNDERGROUND ENGINEERING APPLICATION*

Howard A. Tewes
Lawrence Radiation Laboratory
Livermore, California

ABSTRACT

The constraints on nuclide production are usually very similar in any underground engineering application of nuclear explosives. However, in some applications the end product could be contaminated unless the proper nuclear device is used.

This fact can be illustrated from two underground engineering experiments--Gasbuggy and Sloop. In the Gasbuggy experiment, appreciable tritium has been shown to be present in the gas currently being produced. However, in future gas stimulation applications (as distinct from experiments), a minimum production of tritium by the explosive is desirable since product contamination by this nuclide may place severe limitations on the use of the tritiated gas. In Sloop, where production of copper is the goal of the experiment, product contamination would not be caused by tritium but could result from other nuclides: Thus, gas stimulation could require the use of fission explosives while the lower cost per kiloton of thermonuclear explosives could make them attractive for ore-crushing applications.

Because of this consideration, radionuclide production calculations must be made for both fission and for thermonuclear explosives in the underground environment. Such activation calculations on materials of construction are performed in a manner similar to that described in another paper, but radionuclide production in the environment must be computed using both

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fission neutron and 14-MeV neutron sources in order to treat the "source term" problem realistically.

In making such computations, parameter studies including the effects of environmental temperature, neutron shielding, and rock types have been carried out. Results indicate the importance of carefully evaluating the radionuclide production for each individual underground engineering application.

INTRODUCTION

It is necessary to ascertain the total "source term" (radionuclide production) resulting from the detonation of an underground engineering explosive to allow adequate analysis to be formulated of both the safety and economic aspects of an application. In the realm of safety, although the radioactivity produced is not immediately broadcast to the biosphere, possible accidental seepage to the surface must be considered, as must potential contamination of mobile ground water.

The economic aspects of the application are directly affected in those cases where the product undergoes a refining process of some sort prior to its ultimate utilization; if the raw material contains appreciable quantities of radioactivity, the processing equipment must be designed to prevent unacceptable radiation exposures to the equipment operators.

Finally, the question which impacts on both the safety and economic areas is that of acceptable product contamination: What is the initial concentration of radioactivity and what is the cost of reducing the initial levels of radioactivity to those required by accepted safety standards?

RADIONUCLIDE PRODUCTION

Sources of Radionuclides

Fission Products.--Although the yields of the various fission products may differ appreciably for different fissile materials and are sensitive to the energy of the neutrons initiating the fission, existing studies[1-3] enable adequate estimates to be made of fission product production in Plowshare explosives.

Neutron Activation Products: Device Components.--As presently defined, "device components" include those portions of a Plowshare explosive package which can be subjected to very high time-integrated neutron fluxes. Radionuclide production in these components can be extensive; for sake of completeness, multiple neutron-induced reactions producing radioisotopes far from the stability curve[4] are included in the calculation.

Materials farther away from burning fissile or thermonuclear fuel are exposed to somewhat lower time-integrated neutron fluxes; hence, first-order reactions predominate in the production of radionuclides.

Neutron Activation Products: Canister and Soil.--Activation products formed at relatively large distances from neutron sources will consist primarily of those made by the neutron capture process. Whether neutron shielding material will be employed (as is the case for nuclear excavation explosives) or not, the neutron spectrum incident upon the canister and soil will include some high-energy neutrons, and some (n, 2n), (n, p) and (n, α) reactions will be induced in the canister and adjacent soil. However, the neutron spectrum "softens" rapidly in the shielding and soil or rock (especially where significant water is present), thus causing the overall predominance of (n, γ) reactions.

CALCULATIONS (CODES AND PROCEDURES)

Computation of Neutron Fluxes

High-flux Regions.--Neutronic calculations may be made of the explosion phase of a fission or thermonuclear device using neutron diffusion or Monte Carlo computational techniques. Current versions of computer programs using these techniques not only provide for the calculation of neutron fluxes (divided into a number of energy groups) as a function of time in any region of the device included in the problem, but also allow the calculation of nuclide production from multiple reactions occurring in any of these regions.

Existing codes also provide for the estimation of the total number of neutrons emitted from the outermost region included in the problem, again as a function of time and divided into several energy groups. Thus, these codes produce a "source term" for additional calculations of relatively low neutron fluxes further away from the explosive.

Low-flux Regions.--The most definitive work[5] which has been done to date on obtaining neutron fluxes external to

the explosive involves the use of the neutronic codes mentioned previously. Instead of incorporating only the explosive geometry into the coordinate system used in the calculation, a large segment of the surrounding material (canister and rock, for example) are also considered. Thus, as the nuclear explosion proceeds, the interaction of the surroundings with the explosive is treated, and a more accurate assessment of neutron capture times and neutron energies at the time of capture can be made. Figure 1 shows the temperatures existing around an explosive as a function of time after detonation. Despite the obvious advantages of this computational approach, it can be put to only limited use due to the large amount of computer time required to obtain results. Thus, this method is used only to obtain some guidance on the behavior of specific devices; for subsequent parameter studies (such as activation of different varieties of rock) the neutron output from a conventional explosive calculation is coupled with a Monte Carlo[6] code which is used to estimate time-integrated neutron fluxes in various regions surrounding the explosive. However, by using the appropriate results regarding explosive configuration and environmental temperature obtained from the preceding more rigorous calculations, the simplified procedure can yield valuable and reasonably accurate results in only a fraction of the time required for the more sophisticated calculation.

COMPUTATION OF CROSS SECTIONS FOR NEUTRON-INDUCED REACTIONS

Although a large number of experimentally-determined cross sections and excitation functions are available for neutron-induced reactions (see Refs. 7 and 8), neutron cross sections for those nuclides that are involved in the multiple reactions occurring in high-flux regions are not easily measurable, and are not available at this time. Hence, appropriate codes are under development to calculate these needed cross sections.

The computation of (n, 2n) reaction cross sections, using a normalized statistical model approach,[9] has been generally quite successful. Although there have been no experimental checks of the validity of this cross-section calculational approach on those nuclides far from stability, the use of this model for two or three mass units on either side of the region of stability should give satisfactory results.

Again, for the (n,α) reaction, a combination of statistical and empirical calculations[10,11] seems to provide adequate results. Since charged-particle emission in general competes rather poorly against de-excitation by neutron or

photon emission, the reaction products are not formed in great abundance, and the accurate estimation is not as critical (at least where gross gamma field predictions are concerned) as is the case for the (n,2n) and (n,γ) reactions.

The (n,p) reaction can proceed not only by means of the compound nucleus, but also through charge exchange (direct interaction). Thus, a theoretical treatment of this reaction becomes somewhat involved, and an empirical predictive method[12] is currently being used.

Of critical importance for the accurate calculation of neutron activation is an adequate predictive capability for (n,γ) reactions. Although these reactions have been studied in detail, both for the purposes of reactor and explosive design, as well as in the formulation of cosmological theory, recent evidence[13] indicates that serious gaps exist in nuclear reaction theory which makes extremely uncertain the prediction of (n,γ) cross sections by a theoretical approach. For the present, semi-empirical calculations are being used; however, work is continuing on a more adequate and reliable procedure.

ACTIVATION CALCULATIONS

High Flux Regions

Due to the incidence of multiple reactions in high flux regions, it has been necessary to develop "bookkeeping" codes to keep track of the build-up and depletion of individual nuclides in such regions. Essentially, the codes employ a calculated neutron flux (as obtained from a neutronic code output) at appropriate time intervals during the "burn" of fissile or thermonuclear fuel and, using the required neutron cross sections, calculate the nuclide composition within each region of interest as a function of time. The availability of an extensive nuclide "grid," as well as of a large library of neutron cross sections, allows the calculation of multiple reactions even on multi-isotopic elements. Figure 2 shows the multiplicity of cross sections which must be considered in a calculation of this sort.

Two major codes have been developed for this nuclide-accounting operation. The first, NOVA,[14] was originally written to obtain a predictive capability for heavy-element production in uranium targets subjected to intense neutron irradiation during the detonation of a nuclear device. It has since been rewritten slightly to allow its use with additional target elements.

The second code, ACT,[15] has been designed to utilize more of the neutron flux data generated by the neutronic codes in order to give a more accurate estimate of nuclide production in especially high flux regions. It also has the capability of calculating the radioactive decay of the various product radionuclides as a function of time.

Low Flux Regions

Since only single-order reactions are considered in such regions, a much simpler "accounting" code is required. For instance, there is no need to consider the build-up of nuclides as a function of time. Thus, the output of a Monte Carlo (or extended neutronics) calculation can be coupled with the appropriate compilation of neutron cross sections to obtain the desired list of radioactive species formed.

The ACTIVE code[16,17] has been developed to perform the above-described function; it also calculates the radioactive decay of the radionuclides formed. It has the capability of calculating simultaneously the activation products in all of the regions used in the Monte Carlo calculation and then producing a comprehensive compilation of radionuclides, automatically summing those produced in more than one region.

EXPERIMENTAL CHECKS

Limitations

An obvious test of the adequacy of the predictions obtained above may be made by comparing these results with measured values for radionuclide production. There are two principal difficulties that prevent the simple accomplishment of such a test. First, there are uncertainties in the total chemical composition of an explosive and of its environs. This is especially true of pre-1968 tests, where the need for careful sampling of device materials and soil had not as yet assumed its present importance.

Second, and more important, the chemical fractionation occurring in underground detonations is extreme for some elements, thus making an accurate estimate of the total production of certain radionuclides extremely difficult.[18-21] Consequently, although the calculational check obtained by examining experimental results may be at least semi-quantitative for the so-called refractory elements, data relating to the more volatile elements will probably be relatively unusable for such verification purposes.

RESULTS

Despite the difficulties outlined in the preceding section, some reliable experimental results have been obtained and are given (in a relative way) in Table I. It can be seen that, for the tungsten activation, calculated results are no worse than about a factor of two different from the measured values. It also appears that the predictive capability is improving; i.e., Event No. 3 shows a much better correlation between prediction and measurement than did the preceding Nos. 1 and 2.

Considering the more refractory elements produced mainly in the canister and soil, it can be seen from Table I, that here, too, the predicted production numbers are generally within a factor of two of the observed production.

As more adequate estimates of neutron cross sections become available, it is probable that significant improvement can be achieved in the ability to predict accurately a radionuclide source term.

CALCULATIONAL RESULTS AS RELATED TO UNDERGROUND ENGINEERING APPLICATIONS

Gas Field Stimulation

In the Gasbuggy experiment, a 26-Kt thermonuclear explosive was detonated in a gas-bearing rock; the resulting gas produced from this well contained 18μCi/ft^3 of tritium.[22] This relatively high concentration of total tritium in the produced gas was somewhat lower than the predicted value; however, it emphasizes the need to reduce the tritium production in Plowshare underground engineering explosives which are to be used for gas well stimulation.

Calculations[23] have been carried out for the Gasbuggy Event, allowing the emergent neutrons to impinge on Lewis shale.[24] This work indicates that as much as 1 g of tritium[25] will be formed from the ^{6}Li(n,α)T reaction taking place in the surroundings. Should 30 cm of boric acid be interposed between the neutron source and the shale environment, total tritium production will be reduced by a factor of about 100; about half of this tritium is produced in the soil, while the other half results from the ^{10}B(n,t)2α reaction taking place in the boric acid shielding.

Another source of tritium which cannot be neglected is the ternary fission process; approximately 0.0001 g is formed per kiloton of yield[26] (or a similar amount to that produced in the surroundings by a shielded explosive).

The actual concentration of tritium present in the gas produced from an environment resulting from the detonation of an explosive having such a low tritium yield cannot be specifically assessed; required inputs for a prediction include a detailed analysis of the detonation environment, including the characteristics of the in-place gas.

Ore Crushing

In the Sloop experiment,[27] a nuclear explosive is to be emplaced within a copper-bearing formation and detonated; the crushed ore will subsequently be leached with dilute sulfuric acid to remove and recover the copper.

Preliminary studies have indicated that one of the most troublesome radionuclides, in the sense of being difficult to remove from the copper during processing, is ^{106}Ru. Hence, it would seem that a thermonuclear explosive with relatively little fission would be the most desirable for use in this application. To date, induced radioactivities do not appear to present much of a problem with respect to the copper purification process. Possibly the most significant impact of using a thermonuclear explosive would be the appearance of tritium in the leach solution and hence, in the copper recovery plant. In order to meet occupational safety standards, it might well be necessary to conduct the refining operations in containers which are sealed or appropriately vented to a distant location.

GENERAL CONSIDERATIONS

It can be seen that, on the basis of the preceding instances, each potential Plowshare application must be carefully evaluated with regard to the production and distribution of radioactivity. As a result of this analysis, the appropriate combination of nuclear explosive and shielding will be chosen, the optimum product treatment or recovery plant will be designed, and appropriate research will be initiated to ensure that the product will conform to accepted radiological safety standards.

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Table I. Relative radionuclide production from three Plowshare explosive tests.

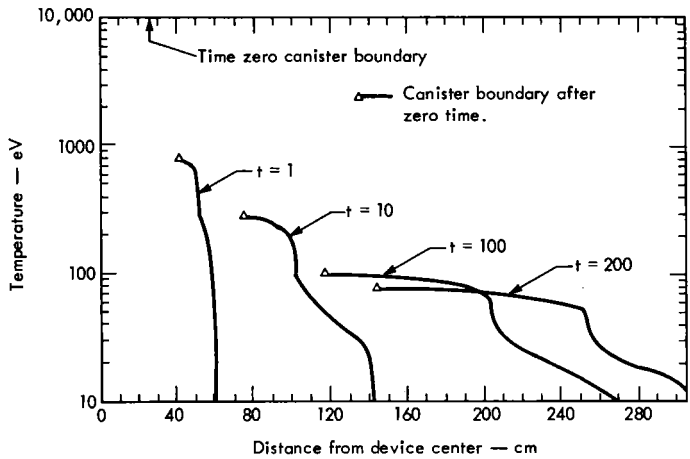
	Atom Ratio: Measured / Predicted		
Nuclide[a]	Event No. 1	Event No. 2	Event No. 3
^{181}W	0.4	0.4	0.8
^{185}W	0.6	0.5	1.1
^{187}W	2.3	0.9	0.7
^{188}W	1.3	1.0	1.1
^{24}Na	1.1	---	---
^{32}P	0.6	---	---
^{51}Cr	0.4	---	---
^{54}Mn	0.7	---	---
^{55}Fe	0.6	---	---
^{59}Fe	0.5	---	---

[a]The tungsten radionuclides were mainly produced in high-flux regions of the explosives; the other radioisotopes were principally formed in lower-flux regions.

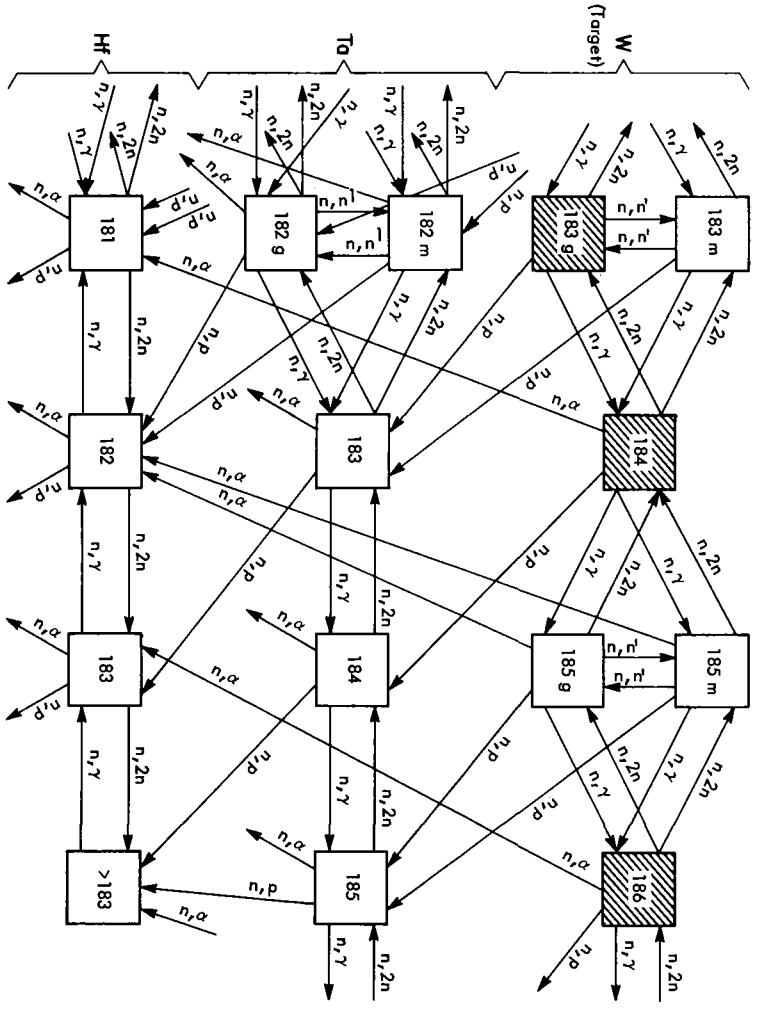
FIGURE CAPTIONS

Fig. 1. Temperature/distance profiles for various relative times after start of nuclear burn, from the neutronic calculations.

Fig. 2. Principal nuclear reactions to be considered in ACT calculations (using a portion of the tungsten nuclides as an illustrative case). Cross-hatched blocks represent stable nuclides; open blocks represent radionuclides.


10,000
1000
100
10
Temperature — eV
Time zero canister boundary
Canister boundary after zero time.
t = 1
t = 10
t = 100
t = 200
0
40
80
120
160
200
240
280
Distance from device center — cm

Tewes - Fig. 1



220-221

Tewes - Fig. 2

QUESTIONS FOR HOWARD TEWES

1. From Charles Bowman:

   With regard to available information, how accurately can krypton-85 and tritium inventories be assessed?

   ANSWER:

   Well, the answer to that is if you are talking about total production, I think that the krypton-85 total production, if you know the fissile material and the neutron spectrum, you could probably get the total production to better than 10%, I would guess. As far as tritium is concerned, that's a little trickier. I would say about a factor of two on that. But in general we try to err on the side of safety. In other words, we will predict it on the high end rather than what we think is the median.

2. From Charles Bowman:

   How accurately can tritium produced from lithium-6 in alpha reactions be predicted?

   ANSWER:

   Well, this is largely a matter of judgment because you see our predictions are just that, we've never really been able to measure this in an explosion environment, I would have to say that, in view of the uncertainties and cross sections and neutron fluxes, I would say like a factor of three.

3. From Charles Bowman:

   What is the critical configuration of lithium-6 about the emplacement point?

   ANSWER:

   The lithium-6 is in shale to the extent of a few parts per million so presumably it's sort of uniformly distributed around a detonation. Of course, if a detonation is taking place in some sort of a layered environment, this might not necessarily hold.

4. From Frank Lowman:

   What would be the fission yield from naturally occurring fissile material in black shale from a 100 kiloton shot based on your calculated neutron fluxes in the soil?

ANSWER:

This depends on the amount of fissile material in the soil naturally. I went through this once, I forget for precisely what yield. I think it was of the order of 100 kilotons and I think the total yield that we were calculating was something of the order of a ton or a few tons. That's tons now not kilotons. That's natural uranium and, I guess, thorium also in the soil.

5. From Antonio Carrea:

Assuming a shot in a foreign country, could an independent safety analysis be done without declassifying information?

ANSWER:

At the present time, no.