

From: Tin Mo *RES*
To: Jason Schaperow
Date: Wed, Feb 16, 2000 4:47 PM
Subject: Re: Importance of Ru to offsite consequences

Jason:

This is to acknowledge receipt of your 2/14/00 e-mail on Ru-106. Before I can answer your questions properly and as accurately as possible with what is currently known about the possible and credible physico-chemical forms (RuO₄ vapor, RuO₂ insoluble particulates) of Ruthenium-106 that are known to be released in accidents, its radiochemistry, biochemistry, metabolism, biokinetics and radiation dosimetry, I need to get more information from you on the following:

(1) What is (or are) a melt down type accident (s) ?

Does your modeling assume that a large amount of heat is released that high enough temperatures are reached to melt the high fired and refractory UO₂ fuel material ?

What is the source of this heat (decay heat only or do you assume in your modeling that the spent fuel assembly went "critical" ?

What is the probability that the fuel assembly would go "critical" under the accident scenario you choose to model?

Is fire involved in your accident scenario ?

What are the highest temperatures reached in your simulation?

(2) If available I would like to review the report on the Canadian experiment to find out if it gives any information on the physico-chemical form(s) of Ru-106 released in their small scale test. That is, is it RuO₄ gas, or is it RuO₂ particulates, or is it RuO₂ incorporated into UO₂ and fuel cladding particulate aerosols ? I supposed the form of Ru-106 released will depend on what actually happened during the postulated (simulated) accident. If the fuel and cladding got heated and melt, it is possible that whatever RuO₄ that exist or is formed by oxidation of RuO₂ in the fuel will vaporized and be released as a gas. RuO₄ is a gas at temperatures above 28 degrees Centigrade but is readily reduced to ruthenium dioxide (RuO₂) in the presence of water vapor and various mildly reducing compounds that one would encounter in fluids in the human respiratory system.

If for some reason (which I do not know since I do not know the details about either the Canadian experiment or your accident simulation/modeling) the fuel broke up into small pieces and there is enough kinetic energy available to disperse it as an aerosol, some of the Ru-106 could be still trapped within the fuel particles and released as such. If inhaled, the metabolism and dosimetry of such inhaled material may be different than that of RuO₄ or RuO₂ and the dose conversion factors may be also different than those in Federal Guidance Report No. 11.

Based on what is published in the open literature for the last 2 decades and on research performed at Lovelace Inhalation Toxicology Research Institute (ITRI) during early 1970 to mid-1980, when I was still working there, the majority of Ru-103 or Ru-106 released in accidents in Japan, U.S., etc. nuclear facilities is determined to be in the form of RuO₄ (gas). Therefore, during 1970 to mid 1980 my colleagues at ITRI, G. Newton, B. Snipes, et al conducted research on inhalation toxicology, biokinetics and radiation dosimetry of Ru-106 using RuO₄ gas. They also conducted inhalation toxicology research using RuO₂ particulate aerosols because industrial accidents in nuclear fuel processing facilities could involve fire which could convert or thermally degrade any ruthenium released to RuO₂. These studies at ITRI supported by U.S.AEC and ERDA indicate that biokinetics (metabolism) and radiation dosimetry and radiological impacts of inhaled Ru-106 released from an accident would depend

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somewhat on what physicochemical form it is initially inhaled by the human receptor or the member of the public.

In your modeling did you assume that all the Ru-106 inventory in the fuel is released ?

I am not too familiar with the dose assessment module of the MACCS code. Is the release source term determined/estimated outside this module and then input into the dose calculation module ? How is this estimation done ? Is it a conservative/ bounding estimate ?

For consequences analysis of most nuclear facilities accidents not involving an operating reactor, historical radionuclide release estimation efforts have produced a consensus that the source term can be defined as a product of five material-specific factors:

$$\text{Source Term} = \text{MAR} \times \text{DR} \times \text{ARF} \times \text{RF} \times \text{LPF} \text{ (NUREG/CR-6410),}$$

where

--MAR is the inventory of the material-at-risk (in your case curies or bequerels of Ru-106,Cs-137, etc.),

-- DR is the damage ratio, or the fraction of MAR actually impacted by a given physical force/stress from a specific event modeled/simulated, **It is usually based on consideration of the physical and chemical form of the MAR, technical analysis of the response of the structures to a given stress, and calculational/judgements of the practical limits of the phenomenon (i.e. energy available from the stress).**

--ARF is the airborne release fraction or the fraction of the impacted material (MAR X DR) that can be suspended (aerosolized) to become available for airborne transport following a specific set of induced physical stresses, such as heating, fire, explosion, etc., occurring during a discrete set of release events (i.e.. an intermittent release). For an event where the radioactive material is released continuously, an Airborne Release Rate, ARR (i.e. the fraction of the total available material being suspended or aerosolized per unit time) is used for the source term calculation.

--RF is the respirable fraction or the fraction of the material-of-concern (RuO₄, RuO₂, CsI, etc.) initially suspended in the air and present as particles that can be inhaled into the human respiratory system. The RF is commonly assumed to include particles with Aerodynamic Equivalent Diameters (AEDs) that are 10 micrometers or less. AED is the diameter of a sphere with a density of 1 g/cubic centimeter that exhibits the same terminal velocity as the particle in question,

--LPF is the fraction of airborne "material-of-concern" that leave the confinement/containment barrier after consideration of depletion mechanisms such as precipitation, settling due to gravity or because of agglomeration or filtration (filter or rubble).

The basic progression of the accident /event simulated/ modeled must be understood to obtain realistic estimates of these parameters for the five-factor formula in NURE/CR-6410 to calculate the source terms used to assess downwind radiological impacts. There are, of course, uncertainties associated with each of these estimated values of parameters and each could have a range or probability distribution.

I am assuming that the MACCS code module for estimation of release source terms or your estimation of the release source terms for Ru-106 and Cs-137 uses similar algorithms/parameters contained in the five factor formula in NUREG/CR-6410.

It is to be expected that if you include or account for Ru-106 release in the modeling of your accident scenario, in addition to the Cs-134/137 release, you will get higher doses than if only Cs is assumed to be released for the following reason:

In your case the MAR in curies or mega-becquerels would have doubled in value due to the assumed presence of equal amounts of Ru-106 and Cs-137 in the spent fuel and airborne release of both Ru-106

and Cs-137, and thus increasing the values of ARF or ARR and the source term released which eventually will then translate into larger doses calculated and therefore, a larger number of early fatalities than with a Cs release alone.

The answer to your question, "Why is Ruthenium's inhalation dose conversion factor for the lungs so much higher than cesium's?" is as follows:

Ru-106 has a half life of 370 days and decays by pure beta-emission with an average energy of 0.013 MeV per disintegration. Its progeny (daughter), Rh-106m, decays to stable Pd-106 with a half life of 30 seconds by beta-emission ($E_{\text{max.}} = 3.54$ MeV and gamma emission (5 different energies ranging from 0.513 to 2.41 MeV and averaging 1.30 MeV).

Cs-137 with a half life of 30 years decays by beta-emission ($E_{\text{max.}} = 0.514$ MeV. Its progeny (daughter) Ba-137m decays mainly (about 85 percent) by x-ray-emission with an average energy of 0.662 MeV).

The much higher (Linear Energy Transfer) LET radiations emitted by the Ru-106/Rh-106m and Rh-106 pair of radionuclides plus the longer residence time in the lungs of these radionuclides in much more insoluble (Class Y) RuO₂ respirable particles would be expected to deposit much more energy or deliver a higher dose to the tissues than the rather soluble chemical compounds of Cs-137 (Class D/Ba-137m (Class W) pair of radionuclides which will be cleared much faster from the lungs to the blood stream than the much more insoluble RuO₂ particles (Class Y). That is why the inhalation dose conversion factor (Sv/Bq inhaled) for Ru-106 would be expected to be so much more higher than that for Cs-137.

Because of its basic chemistry (NAS-NS 3029, 1961) and biochemistry (Koda 1970) it is expected that any inhaled gaseous or soluble form of Ru-106 would be rapidly reduced to the dioxide (RuO₂ form) form upon contact with organic compounds found in the lungs and the rest of the respiratory tract. Based upon the results and findings of many years of research on the metabolism and dosimetry of Ru-103 and Ru-106 compounds involving people who accidentally inhaled these radionuclides and inhalation toxicology studies involving animals at ITRI, the ICRP Task Group on Lung Dynamics (TGLD, 1966) assigned oxides and hydroxides of ruthenium (Ru) to inhalation Class Y, halides to Class W and all other compounds to class D. This was endorsed by ICRP in *Publication 30*, Part 2 (1980), in which the assignment of ruthenium dioxide to class Y was supported by experiments with dogs (Stuart, 1970).

For dose assessments, NRC has adopted the dose coefficients presented in Federal Guidance Report No. 11 which are based on the methodology, approaches and recommendations in ICRP *Publication 30*. (I assume that the MACCS code and your dose analysis for your modeling is using the same dose coefficients).

Unless, you can justify using a technically defensible basis to classify Ru inhaled into the lungs as Class W or D, I am afraid you are pretty much stuck with having to use the inhalation Class Y for your dose and health effects modeling. The fact that you assumed or implied that the accident scenario you are trying to model involved melting and oxidation of the fuel cladding will not help you because previous studies at ITRI (Newton and Latven, 1971) had shown that various compounds of ruthenium form RuO₂ when subjected to temperatures above 500 degrees centigrade. If fuel melting is occurring in your accident scenario the temperatures are probably much higher than 1100 degrees centigrade. Even if it was gaseous RuO₄ that was released it will quickly get reduced to very insoluble RuO₂.

I hope this reply answer some of your questions.

I can be reached at 415-8151 or Room T-9 J 10 if you have any questions or want to discuss about my response.

Regards.

Tin

>>> Jason Schaperow 02/14 2:17 PM >>>

I spoke with your acting Branch Chief, Vincent Holahan, and he said you might be able to help me with a question I have.

I am working on analyses of source terms and consequences of accidents involving prolonged loss of water from a spent fuel pool (melt-down type accidents). As a result of radioactive decay from storage in the spent fuel pool, noble gases and iodine have decayed and the only element that is released from the fuel in significant amounts is cesium. However, the Canadians have small-scale experimental results showing that, in an air environment, ruthenium can be released following nearly complete oxidation of the fuel cladding. When I perform a MACCS calculation that includes a ruthenium release, I get a larger number of early fatalities than with a cesium release alone. Because the spent fuel pool I am modeling has about the same number of curies of ruthenium and cesium, the larger number of early fatalities is caused by ruthenium-106's larger inhalation dose conversion factor for the lungs. Why is ruthenium's inhalation dose conversion factor for the lungs so much higher than cesium's?

Also, I noticed that the clearance class (clearance from the lung to the blood) that the ruthenium inhalation dose conversion factor for the lungs in MACCS is based on is "Y," indicating that it takes on the order of years to be cleared from the lungs. Is it appropriate to use this dose conversion factor for calculating early fatalities? (The ruthenium dose conversion factors for the day and week clearance classes are smaller than for the year clearance class.)

Thank you.
Jason Schaperow

CC: ACC, CAT1, CXD, EVH