File: TMI-2
Xenon

MICHIGAN STATE UNIVERSITY

COLLEGE OF NATURAL SCIENCE · DEPARTMENT OF PHYSICS EAST LANSING · MICHIGAN · 48824

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The Honorable Victor Gilinsky
Commissioner
U.S. Nuclear Regulatory Commission
1717 H Street, N.W.
Washington, D.C. 20555

Dear Dr. Gilinsky:

Here is my report in response to the questions you asked me to look into concerning Xe^{133} emissions from the accident at the Three Mile Island Nuclear Station. In order to get this information I had discussions with many scientists and engineers mostly of NRC, but also with others to whom I was referred. I also read relevant NRC reports and other related material. What I've tried to do is to synthesize this information into a coherent picture of the problem of Xe^{133} emission.

The report is ordered under three headings each of which is a question or a few questions.

I. How much Xe^{133} was emitted as a result of the TMI accident? How are the estimates of Xe^{133} emission made? How reliable are the estimates?

The direct answer to these questions is that at least three independent estimates of total Xe^{133} emissions have been made by people. The estimates are 2×10^6, 2.4×10^6, and 13×10^6 Ci. The estimates are rough ones at best, i.e. they make use of rough approximations, and are probably only reliable to within factors of two or three. However, they represent responsible sensible work and are probably the best estimates one can make under the circumstances.

Background: Xenon-133 is produced in the fuel of a reactor by fission of U^{235} during normal operation. The TMI reactor had run for 140 equivalent full power days at the time of the accident and the total amount of Xe^{133} in the

inventory at that time was 140 x 10^6 Ci. This number comes from the origin code, a computer calculation of the radioactive isotopes present as a result of production minus decay, and is generally believed to be reliable. If all of this Xe^{133} were emitted at the beginning of the accident we'd get the maximum amount of Xe^{133} emitted, i.e. an upper bound is 140 x 10^6 Ci.

We have good evidence from DOE environmental samples (helicopter and ground surveys) that the main radionuclide emitted during the accident was Xe^{133}. That means that some Xe^{133} got free from the fuel elements and into the air, probably by the pathway: fuel elements to coolant water, then out of the reactor building into the auxiliary building, then through the filters and into the air via the stack.

During normal operation the emissions from the reactor are measured at the stack by a combination of batch spectral analysis, which measures the relative amounts of the various radionuclides, and a stack monitor, which measures the gross number of curies being emitted. Unfortunately the stack monitor went off scale about two or three hours after the accident started and it stayed off scale. Had that not happened, i.e. had there been a suitably high intensity monitor in the stack, we could have used the stack monitor to estimate the total emissions. Since this can't be done we have to use more indirect, less reliable means. I found out about three of these and they are described briefly with their results below.

A. Estimate from measured doses and meteorological data.

This estimate is described in NUREG-0558 and I discussed it in some detail with Dr. F.J. Congel of the NRC Radiological Impact Section.

Essentially what was done here was to take the doses as measured by thermoluminescent dosimeters in place before and during the accident and to combine these doses with meteorological dispersion factors to get estimates of the strength of the source of the radiation. This gives an estimate of 13 x 10^6 Ci of Xe^{133}

emitted.

The raw dosimeter data for this estimate come from the Teledyne Isotopes dosimeters in place at 20 onsite and offsite locations. The raw meteorological data come from a tower, on the island, which reads wind magnitude and speed and the air temperature at various elevations. These kinds of data are gathered routinely. The meteorological data is analyzed by a meteorology group and they calculate tables of χ/Q, the meteorological dispersion factor. This factor is the ratio of the steady state radioactive concentration at a point in the field (χ in Ci/m^3) to the steady state rate of emission of radioactivity from the stack source (Q in Ci/sec). Thus the ratio χ/Q is kind of a response function, similar to Green's functions used in formal physics. These factors depend in general on direction and distance from the source.

Probably the main source of uncertainty in this estimate of Xe^{133} emission is due to meteorological factors. In general values of χ/Q are thought to be good to a factor of two or three out to a few miles; beyond that the accuracy is worse. At TMI the uncertainty was fairly large due to light winds. Another potential source of uncertainty is in determining the doses to which the TLD's have been exposed. However the National Bureau of Standards has made a preliminary study of the calibrations and that appears to be O.K. (i.e. the uncertainty is about +25% and -30%).

My conclusion is that this value (13×10^6 Ci of total Xe^{133} emission) may be off by a factor of two or three mainly due to meteorological uncertainties.

B. Estimate from delayed grab samples.

This estimate was made by Dr. Andrew P. Hull of Brookhaven National Laboratory and the work was reported at a recent Health Physics Society Meeting.

He obtained data on the actual Xe^{133} concentrations in about five grab samples of the effluent taken at the stack. The samples were taken between April 6-10 several days after the accident, which was on March 28. He plotted these data as

a function of time and extrapolated back to t = 0 using a straight line fit. He then summed up the total Xe^{133} emission and got the value 2 x 10^6 Ci.

The main strength of this method is that it relies on direct measurements of Xe^{133} coming from the stack. The main weaknesses are: first, as mentioned above, the data were taken well after the accident and second, there is probably no real reason to believe that a straight line extrapolation back to the time origin is an accurate representation of what actually happened.

C. Estimate from a remote monitor.

This estimate was described to me by Dr. Carol D. Berger, a health physicist at Oak Ridge National Laboratory. It is independent of the previous estimates and gave a value of 2.4 x 10^6 Ci of Xe^{133} emitted.

Dr. Berger's technique was to use readings from a monitor, in the auxiliary building, that saw from a distance of 40 feet a duct that feeds the stack. This monitor saw part of what went up the stack all the time and it never went off scale, as did the main stack monitor. She calibrated the remote monitor against the main monitor during the short time that the main monitor was still on scale. Associated with this calibration is an uncertainty of a factor of two, but there are other indications that the calibration may have been much better than that. There also is some uncertainty introduced into this method by the necessity of assuming the distribution of radioactive gases in the effluent. This distribution was taken from the inventory at the moment of shutdown, a reasonable choice.

In some ways this is, in principle, the most reliable of the available estimates.

D. I learned in the course of this work that Keith Woodard, a consultant of the utility, estimated the total Xe^{133} emission to be 15 - 20 x 10^6 Ci. I unfortunately do not know what information he used as a basis for his estimate.

II. Could we determine the total Xe^{133} emission from TMI in other ways?

I know of two other ways which in principle might be used to determine

the Xe^{133} emission. One way depends on conservation of Xe and the other depends on Xe which is adsorbed on the filters that precede the stack.

A. Determining total Xe^{133} emitted by conservation of Xe^{133}.

We know from the core inventory at the time of the accident that there were initially 140 x 10^6 Ci of Xe^{133} present. If no more fission or production of Xe^{133} occurred after this time, then the total amount of Xe^{133} present at a time Δt (days) after the accident is 140 x 10^6 $e^{-\lambda \Delta t}$ Ci, where λ = 0.13153 day $^{-1}$ (Xe^{133} has a half-life of 5.27 days). The idea is to measure the total Xe^{133} still in the reactor (containment volume, core, coolant water) and compare this with the above. Whatever is missing has escaped out of the reactor into the environment.

The difficulties with making this work, are: (a) Xe^{133} has a short half-life so that measurements must be made shortly after the accident before it has decayed away to sub-threshold amounts; even then it is a difficult measurement, and (b) It is necessary to measure radiation from Xe^{133} within the reactor where there is so much other radiation present.

As a practical example: Suppose for illustration that 20 x 10^6 Ci of Xe^{133} had escaped at the time of the accident and we were thus left with 120 x 10^6 Ci of Xe^{133} contained on March 28. By August 20, when the present study was undertaken, that would have decayed to 120 x 10^6 $e^{-\lambda 145}$ = 0.63 Ci still contained within the reactor if no more had escaped. This means, in order to detect whether any Xe escaped initially it would be necessary, by August 20, to measure the total Xe^{133} present to better than about $\pm$ 0.1 Ci out of a total of 0.6 Ci all against the large background of other radionuclides in the reactor. That's probably much too hard to do.

There is an interesting application of this principle to the TMI accident, and an associated puzzle. On June 29, NRC engineers measured a Xe^{133} concentration of 1.5 x 10^{-2} μCi/ml in the gas space of the containment volume. The volume of this gas space is 2 x 10^6 cubic feet, the volume of water in the containment vessel is 525,000 gallons, the solubility of Xe in water is about 0.1,

and Δt between 3/28 and 6/29 is 93 days. From these data I calculate that on 6/29 there were present about 850 Ci of Xe^{133} in the gas containment volume, and a negligible 3 Ci of Xe^{133} in the water volume. This means that, if we assume that no Xe^{133} escaped or was added to the containment building after the accident, there were about 850 $e^{+\lambda 93}$ = 170 x 10^6 Ci of Xe^{133} present initially. That implies that essentially all of the Xe^{133} escaped from the fuel into the containment building. In fact 170 x 10^6 Ci is even somewhat larger than the origin code value. The puzzle is that working from this same data, NRC engineers have concluded that half the noble gas was released from the fuel rods. I do not understand the discrepancy between our conclusions.

B. Determining total Xe^{133} emitted by analysis of filters.

The idea here is that all of the Xe^{133} that comes out of the stack first passes through HEPA filters and charcoal filters. The charcoal filters are 2" thick beds of carbon and some of the Xe remains on the charcoal filters. The mechanism for this is probably surface adsorption of Xe and subsequent diffusion into the bulk carbon.

In principle it might be possible to tell something about how much Xe^{133} passed through the filters from measurements of how much Xe^{133} they retained. However I believe that this cannot with present knowledge tell us anything quantitatively useful about the Xe^{133} emission. The difficulty is that we would have to understand the interaction between Xe and carbon very well, then we'd have to know the entire history of the filters since they were exposed to Xe: Did they ever get wet? Did fresh air blow over them? Did they ever get warm? etc. These are the kinds of things that determine the connection between how much Xe^{133} the filters contain now and how much Xe^{133} passed through them.

I understand from Mr. John T. Collins that some of his group of engineers are working with the charcoal filters from the TMI - 2 reactor.

III. What can be done so that Xe^{133} emissions and doses will be measured reliably in any future accidents?

In retrospect the best single way to insure that these emissions are reliably monitored is to install a stack monitor that will not go off scale during high radioactivity emissions. This may require a series of monitors which work from low concentrations to high ones with overlapping ranges.

There is a closely related question: How can the population doses due to Xe^{133} emissions be measured more accurately? This can probably most conveniently be done by increasing the number of TLD's that are routinely in place, by more dense distribution of their locations, and by using TLD's that are well calibrated for the gamma (81 keV) and beta (350 keV) emissions of Xe^{133}.

IV. How can Xe^{133} emission be prevented in any future accident?

This is difficult to answer optimally since the pathway for the emission isn't yet clear. However, the following may work: Xe has boiling point and triple point temperatures of, respectively, $165^{o}K$ and $161^{o}K$. Probably if one provided surfaces which were cooled to liquid nitrogen temperatures and over which the Xe^{133} had to pass in close proximity, then a large fraction of the Xe could be condensed and held until it decayed. The same kind of condensing plates would probably work with Kr^{85} whose boiling point and triple point temperatures are, respectively, $120^{o}K$ and $116^{o}K$. There are a few practical problems in implementing this but the method should be straightforward. For example it would probably suffice just to cool charcoal filters when the Xe^{133} emissions were high and let the Xe be condensed and retained on them.

Report submitted by

Gerald L. Pollack

Gerald L. Pollack

Professor, Physics Department