Preliminary Analysis of the Criticality Accident at Tokaimura

DRAFT II

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While we are unlikely to know precisely what happened at Tokaimura for weeks to come, sufficient information has become available to put the accident in perspective. Tokaimura, about 110 kilometers (70 miles) northeast of Tokyo, is the site of Japan's spent fuel reprocessing plant. It appears that the accident occurred in uranium fuel processing plant at the site—the JCO Co. Ltd. Conversion Test Building—and involved recycling scrap enriched-uranium dioxide (UO_2) recovered from the manufacture of fresh fuel for the 100 MW Joyo research fast breeder reactor. Joyo is designed to operate with 18% enriched uranium, with a minimum critical mass of 46 kilograms (kg). The enrichment of the UO_2 scrap was reported to be 18.9% enriched in the isotope uranium-235 (U-235).

It has been reported that workers caused a criticality accident, an uncontrolled chain reaction, when they placed 16 kg, instead of 2.4 kg, of enriched uranium in a tank of nitric acid. The tank was surrounded by a water jacket. The reports appear to indicate that the criticality was reoccurring—the chain reaction occurred continuously or intermittently—and lasted for about 19 hours and 40 minutes. It started at 10:35 am local time (0135 GMT) on September 30, 1999, and was not brought under control until the water jacket was drained at 6:15 am the next morning. They later added boron to the tank for added safety. Apparently, the workers created what in effect was a solution reactor with a water reflector.

There have been 22 out-of-reactor criticality events in the United States.[1] Twenty-one events occurred between 1945 and July 24, 1964. The only other U.S. criticality accident occurred in October 1978, and involved a solvent-extraction process at the Idaho Chemical Processing Plant.[2] In May 1991, at a General Electric plant at Wilmington, North Carolina, there was a transfer of low-enriched uranium to a large waste tank that resulted in conditions favorable for a criticality event, but no criticality accident resulted.[3]

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[1] *The Technology of Nuclear Reactor Safety*, Volume 1, T.J. Thomson and J.G. Beckerley, editors (Cambridge: The M.I.T. Press, 1964), Table 2-1; and D.W. Croucher, Chairman, Nuclear Criticality Safety Committee, "Criticality Safety Considerations Pertinent to Transition," EG&G Rocky Flats, March 1993, viewgraphs.

[2] Croucher, "Criticality Safety Considerations Pertinent to Transition."

[3] Ibid.

Of the 22 criticality events and one near miss in the United States, only seven involved chemical processing of enriched-fissile materials. Of these, three or four events resulted in reoccurring criticality. The total number of fissions occurring during the three largest of these out-of-reactor criticality events ranged from about $6x10^{17}$ to about $4x10^{19}$. The out-or-reactor criticality event in the United States that resulted in the largest energy release, i.e., the most fissions ($4x10^{19}$), occurred on October 16, 1959, at the Chemical Processing Plant at NRTS (now called the Idaho National Environmental Engineering Laboratory (INEEL). This event involved repeated excursions and boiling. The event with the longest period of reoccurring criticality, 37 hours, began on April, 7, 1962, at one of the chemical processing plants at Hanford and resulted in about $8.2x10^{17}$ fissions.[4]

A U.S. analog to the Tokaimura accident occurred at the Wood River Junction, Rhode Island, Scrap Recovery Plant on 24 July 1964. In this accident, concentrated uranyl nitrate solution [$U^{235}O_2(NO_3)_2$ soln.] was hand-poured into a geometrically unsafe container.[5] The result was about $1x10^{17}$.[6] It is unclear whether this was a reoccurring criticality.

Each fission releases about $3.2x10^{-11}$ joules.[7] Therefore, typical reoccurring criticality events involving chemical processing operations, resulting in $6x10^{17}$ to $4x10^{19}$ fissions, released 19 million to 1.3 billion joules.

The Tokaimura reoccurring criticality lasted for up to $19^2/_3$ hours. The average power level during this period was probably not more than a few kilowatts, otherwise the reaction probably would have shut down sooner as a consequence of the nitric acid solution boiling away. Therefore, the total energy released was probably not more than a few hundred million joules, or in the range of the historical values.[8]

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[4] *The Technology of Nuclear Reactor Safety*, Volume 1, Table 2-1.

[5] The plant was designed to recover HEU from unirradiated scrap material produced during fabrication of reactor fuel elements. Uranium-contaminated trichloroethane solution with very low concentrations of uranium was recovered by mixing with sodium carbonate. Because of the large volumes of solution, the operation was being conducted in a makeup tank about 18 inches in diameter and 25 inches deep--an unsafe geometry for concentrated solutions. (The operation was supposed to be performed in small, criticality safe, bottles but this procedure had been bypassed to deal with a large amount of solution.) The day before the accident, an evaporator failed to operate properly and a plug of uranium nitrate crystals was created. These were dissolved with steam and the solution (240 g U-235/cm3) was drained into polyethylene bottles identical to those used for the trichloroethane solution. A worker confused the bottles and poured concentrated solution into the makeup tank. USAEC, LA-3611, September 22, 1967.

[6] USAEC, LA-3611, September 22, 1967. *The Technology of Nuclear Reactor Safety*, Volume 1, Table 2-1 gives the worker dose as ">700 rem (fatal)."

[7] Assumes 200 MeV per fission.

[8] Assuming one kilowatt: (1000 W)(19.67h)(3600s/h)(1joule/W-s) = $7x10^7$ joules. This corresponds to $2.21x10^{18}$ total fissions.

A modern 1000 Megawatt-electric (MWe) nuclear power plant produces about 3400 Megawatts of thermal energy. If one assumes that the Tokaimura event averaged 3.4 kilowatts during the 19.67 hours, the rate of fissions during the Tokaimura event would be one million times less than that in the 1000 MWe power plant. Consequently, the rate of production of the various fission products also would be about one million times less.

The Tokiamura event did not result in an explosion like a nuclear weapon since the fissions occurred over a period of hours instead of microseconds. Nevertheless, it is possible to compare the total fissions to demonstrate that the fission products generated during the Tokaimura event were in no way comparable to those generated and released during a nuclear weapon explosion. At Tokaimura the probable upper range of total fissions was $6x10^{17}$ to $4x10^{19}$. This is 3600 to 240,000 times less than the $1.45x10^{23}$ fissions in a one kiloton nuclear weapon.

The first two criticality events that occurred at Los Alamos on August 21, 1945 and May 21, 1946, each resulted in the death of a worker in the room where the criticality event occurred. The first event involved an estimated $3x10^{15}$ fissions and resulted in a fatal prompt radiation dose estimated to be 800 Roentgens (R). The second involved about 10^{16} fissions and resulted in about 900 R.[9] The 1964 Wood River Junction criticality that resulted in $1x10^{17}$ fissions exposed a worker to about 10,000 rad.[10] He died 49 hours later. The LD_{50}, the acute dose which is fatal to 50 percent of the population, is 375 to 475 R, assuming no medical intervention.[11] Three workers at Tokaimura reportedly received high doses; two were reported to be in critical condition; and one is said to have had diarrhea, a symptom of acute radiation sickness. Thus, absent heroic medicine, and perhaps in spite of it, one or two workers have a high probability of dying within one or two months.

In addition to the exposure of workers nearby to prompt radiation, it appears that radioactive fission products leaked from the plant as evidence of the 70-85 millirem dose measures at the site boundary during the first 24 hours of the accident. A primary at-risk population is children, and a key concern is their dose to the thyroid due to exposure to radioiodine. In this case the three most important isotopes of iodine are iodine-131 (I-131), which has a half-life of 8 days ($T_{1/2}$=8.04 days), I-133 ($T_{1/2}$=20.8 hours) and I-135 ($T_{1/2}$=6.55 hours). Approximately 2.26 percent of the thermal-neutron fissions of U-235

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[9] *The Technology of Nuclear Reactor Safety*, Volume 1, Table 2-1.

[10] USAEC, LA-3611, September 22, 1967.

[11] *The Effects of Nuclear Weapons*, Samuel Glasstone, Editor (Washington, D.C.: United States Atomic Energy commission, June 1957), Figure 11.57, p. 470.

results in a I-131 atom—4.36 percent for I-133 and 2.29 percent for I-135.[12] Therefore, $6x10^{17}$ to $4x10^{19}$ fissions would result in $1.4x10^{16}$ to $9x10^{17}$ atoms of I-131, which represents 0.4 to 24 curies (Ci) of I-131, and 20-1200 Ci of the three iodine isotopes.[13]

By comparison the core of the Three Mile Island Unit 2 reactor (TMI-2) had an estimated 70 million Ci of I-131 in the reactor core at the time of the accident.[14] During the period from 3 to 10 hours, an estimated 2.6 percent of the I-131 inventory was vented into the secondary containment building, and ultimately an estimated 12 percent of the I-131 was assumed to have entered the TMI-2 secondary containment.[15] Less than 0.00004 percent of the I-131 inventory, i.e., less than about 30 Ci, was believed to have been released into the environment.[16] The Tokaimura plant did not have a secondary containment. How much of the radioiodine and other fission products escaped during the Tokaimura accident is unknown, but it is unlikely to be significantly greater than that released to the environment from TMI-2.

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[12] K.A. Varteressian and Leslie Burris, "Fission product Spectra from Fast and thermal Fission of U^{235} and Pu^{239}, Argonne National Laboratory, ANL-7678, March 1970, table C.29, p. 314; The I-131 fraction assumes one day irradiation time.

[13] Assuming a cooling period (i.e., travel time to the point of inhalation) of 15 minutes does not reduce the activity of these isotopes significantly.

[14] *The Three Mile Island Accident*, L.M. Troth, A.P. Malinauskas, G.R. Eidam, and H.M. Burton, Editors (Washington, D.C.: American Chemical Society, 1986), p. 46.

[15] Ibid., p. 281.

[16] Ibid., p. 48.