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Criticality accident at Tokai nuclear fuel plant (Japan)"

What happened?

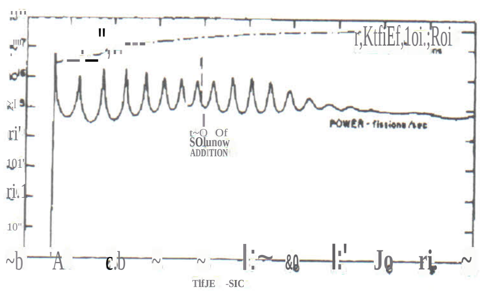
On September 30, 1999, a criticality accident occured at the Tokai nuclear fuel plant in Japan. The plant, operated by *lCO* Co. Ltd., a 100% subsidiary of Sumitomo Metal Mining Co. Ltd., converts enriched uranium hexafluoride (UF_6) to uranium dioxide (UO_2) for use in nuclear fuel. Criticality accidents involve a self-sustaining chain reaction caused from handling of too large amounts of enriched uranium. The chain reaction continued for around 20 hours, before it could be stopped.

The uranium processed was enriched to 18.8% of uranium-235, rather than the 3-5% used for commercial light water reactor fuel. Material of this high enrichment grade was being produced for the experimental *loyo* Fast Breeder reactor.

The plant has an annual production capacity of 715 tonnes of uranium for light water reactors, and 3 tOIlI1eS of uranium for fast breeder reactors.

The conversion process used in the Tokai plant is a wet chemical process: The uranium oxide (U_3O_8) obtained in a first processing step (in the form of a powder) is dissolved in nitric acid (HNO_3). The resulting uranyl nitrate solution or slurry ($UO_2(NO_3)_2$) is pumped into a buffer tank. Then, the liquid is pumped into a precipitation tank, where anunonia (NH_3) is added to precipitate anunonium diuranate (~)2U207 ("ADU"). The amount of uranium contained in this tank must be strictly controlled to avoid criticality. These controls however failed, and 16 kg instead of the permitted 2.4 kg were poured into the tank. About 6 kg were already sufficient to initiate a criticality under these circumstances.

In dry systems, criticality accidents often are quenched by various inherent effects, terminating the chain reaction automatically. In solution criticality accidents such as in Tokai however, the chain reaction may continue for some time after the first high peak and typically is followed by fissions of oscillating intensity, see til(following graph:



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(from McLaughlin_1991)

The Tokai precipitation tank was equipped with a water-filled cooling jacket. After the chain reaction was initiated, this water possibly' contributed to the effect that the reaction did not stop automatically after the first peak. Water is a good reflector and moderator to slow down neutrons for they can initiate further fissions of uranium-235 atoms. The chain reaction only stopped, after this water had been drained and boron (a neutron absorber) had been added. Based on the results from analyzing the sample

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solution taken form the precipitation basin on October 20, 1999, the STA assumes that the entire nuclear fission number resulting from the criticality reaction is 2.5 · 10 18, and the weight of uranium-235 that underwent fission was about 1 mg. Nearly half of the total fissions (1.2 · 1018) apparently occured during the first 25 minutes of the criticality. The total energy generated during the criticality event was estimated to have been about 22.5 kWh.

Impacts

The chain reaction caused heavy releases of gamma and neutron radiation. Three workers were exposed to doses of up to 17 Sv (Sieverts), causing severe radiation sickness. The worker exposed to the higest dose died on December 21, 1999. The worker exposed to the second highest dose of 6 - 10 Sv died on April 27, 2000. 68 other persons were irradiated at lower levels. Among them were the workers who stopped the chain reaction: they were exposed to doses of up to 119.79 mSv, exceeding the 100 mSv limit for emergency situations. The annual dose limit for workers is 50 mSv (while ICRP currently recommends 20 mSv). As of October 7, 1999, radiation levels remained high inside the plant building, preventing inspection of the damage inside the plant. First reports that an explosion had blown a hole into the roof of the facility have not been confirmed.

On November 4, 1999, the Science and Technology Agency submitted a report to the Nuclear Safety Commission, containing estimates of radiation exposures during the criticality accident, based on an analysis of uranium solution sampled from a tank inside the fuel processing plant and neutron levels monitored around the plant. The report says that the radiation dose received at a distance of 80 meters from the accident site (that is the nearest boundary of the plant) was estimated at 75 mSv(this figure was revised to 30 mSv, according to Yomiuri Shimbun of Dec. 12, 1999. Details are not known yet.) for the first 25 minutes of the criticality accident, and 160 mSv for the whole criticality period of 20 hours; the radiation dose received at a distance of 350 meters from the plant (that was the evacuation boundary), was estimated at almost equal to the annual permissible dose of 1 mSv for the first 25 minutes, and 2 mSv for the whole event.

Since the evacuation started only 5 hours after the begin of the criticality, residents may have received doses of more than 75 mSv, that is 75 times the permissible annual dose of 1 mSv. The government has so far said 69 people were exposed to radiation, but the latest survey says the number of affected people could increase.

On January 31, 2000, the Science and Technology Agency reported that the JCO Co. nuclear accident exposed 119 people to levels of radiation that were higher than the maximum dose of one millisievert, considered safe f-11 an ordinary person to be exposed to in a period of one year. The agency checked radiation doses received by 207 people living within a radius of 350 meters from the accident site, 148 JCO employees who were on its premises at the time of the accident, 60 firefighters and government officials who attended the scene, and 24 JCO technicians who entered the plant in an attempt to halt the self-sustaining nuclear reaction of a uranium solution. Among local residents, the highest radiation dose recorded was 21 millisieverts. The recipient lived right next to the plant and took a long time to evacuate, the agency said. (Yomiuri Feb. 1, 2000)

The estimate of the total number of persons exposed to radiation from the accident was raised to 439. Of those affected, 207 were local people who lived and worked within a 350 m radius of the plant. (Reuters Jan 31, 2000). On Oct. 14, 2000, the Science and Technology Agency raised the number of people exposed to radiation to 667. The number includes 229 people - passers-by, government officials and those from new organizations - who have newly been found to have been exposed to radiation in the accident. Of the 229, 56 were overexposed, registering more than 1 millisievert of radiation. One person registered 7.2 millisieverts. (Mainichi Oct. 15, 2000).

During the accident monitoring of neutron and gamma dose rates started only several hours after the begin of the chain reaction. Since recent analysis shows that about half of the fissions occured during the first 25 minutes, the significance of such delayed monitoring is very limited: At the boundary of the facility, gamma dose rates of 0.84 mSv/h and neutron dose rates of 4.5 mSv/h were recorded, while the dose limit for members of the public is 1 mSv per year.
Two minutes after the accident, neutron radiation levels of 0.26 micro-Sv/h, that is 10 to 100 times the normal level, were detected at a laboratory of the Japan Atomic Energy Research Institute in Nakamachi, located about two kilometers from the JCO plant.

While gamma radiation is hazardous only while it is being emitted, neutron radiation produces new radioactive isotopes in irradiated matter - it activates the matter. These newly formed isotopes, such as sodium-24 and zinc-65, continue to emit radiation at their respective half-lives, even after the neutron radiation has stopped. On the one hand, this effect is a matter of concern for the residents living nearby the plant, on the other hand it can be used to estimate the neutron dose rates received at such places, where no neutron monitoring had been performed.

Personal radiological data calculated from whole-body counting of Na-24 is available from STA:

- JCO workers #1-20; #21-42 : up to 23 mGy (3 most severely irradiated workers not included)
- 3 Fire fighters: up to 3.9 mGy
- 7 Area Residents: up to 9.1 mGy

The chain reaction also produced gazeous fission products, such as radioactive isotopes of iodine, krypton, and xenon. These, as well as radioactive cesium, probably have been released into the environment. The following is an inventory of fission products detected within 3 km of the accident site. The figures were taken from newspapers and TV reports, and have been compiled by CNIC:

- strontium-91 --- 0.021 *Bq/m3* in air, 900m southeast of the site
- strontium-91 (krypton-91) --- unknown amount, location not specified
- iodine-131 --- 54.7 Bq/kg from mugwort leaves, 100m from the site
- iodine-133 (krypton-91) --- unreported amount, 100m from the site
- cesium-137 --- unreported amount, 7 locations
- sodium-24 --- 64 Bq/kg, 300m west from the site
- sodium-24 --- 1.7 Bq/kg, 3km west from the site
- xenon-13 9 --- from the vomit of the exposed workers
- krypton-91 --- from the vomit of the exposed workers

The accident has been rated Level 4 on IAEA's 7-level International Nuclear Event Scale (INES) .

On Oct II, 1UOO, six former executives and current employees of JCO Co. were arrested on "Suspicion of negligence resulting in two employees' deaths. Ibaraki Prefectural Police spent more than one year investigating the nuclear accident and have apparently concluded that JCO's systematic rule violations and a lack of safety measures jointly caused the disaster. Ibaraki police arrested Kenzo Koshijima, 54, who was the head of the JCO Tokai plant where the accident took place on Sept. 30, and five other officials of the company. (Mainichi Shimbun Oct. 12,2000)

Six former top officials at JCO Co. pleaded guilty to charges of negligence resulting in death as the trial opened into Japan's worst nuclear accident. The pleas were entered at the initial hearing at the Mito District Court in Ibaraki. Also, current JCO Co. president Tomoyuki Inami admitted to a charge that the company violated the nation's nuclear regulations law, said Mito District Court spokesman Michiru Sakurai. (Reuters Apr 23, 2001)

Criticality accidents worldwide

A total of about 60 criticality accidents has been reported worldwide. Most of them occured in the 1940's to 1970's in nuclear (weapons) research laboratories.

"In the United States there has been only one incident in which criticality was reached at a commercial nuclear fuel processing plant. That occurred in 1964 at a plant, no longer in operation, near Charlestown, Rhode Island. A worker accidentally set off the reaction when he poured liquid uranium into a tank and died after being exposed to 1,000 times the lethal dose of radiation." (San Jose Mercury News, Oct. 1, 1999)

A near-criticality incident occured on May *28129,* 1991, at General Electric's Wilmington (North Carolina) nuclear fuel fabrication facility. Approximately 150 kilograms of uranium were inadvertently transferred from safe process tanks to an unsafe tank located at the waste treatment facility, thus creating the potential for a localized criticality safety problem. The excess uranium was ultimately safely recovered when the tank contents were centrifuged to remove the uranium-bearing material. (for details, see: Potential Criticality Accident at the General Electric Nuclear Fuel and Component Manufacturing Facility, May 29, 1991, Investigation Report, Nuclear Regulatory Commission, Washington, DC., NUREG-1450, August 1991,227 p.)

- Minor incidents affecting criticality safety are occuring quite often. Recent examples:
- October 1, 1999: BWX Technologies Lynchburg, Virginia
- August 13, 1999: Siemes Power Corporation, Richland, Washington
- July 2, 1999: Portsmouth Gaseous Diffusion Plant, Ohio
- May 18, 1999: BWX Technologies Lynchburg, Virginia

The last criticality accident reported from the former Soviet Union occured on June 17, 1997 in the Arzamas-16 nuclear weapons laboratory. One researcher died.

Reference

Process criticality accident likelihoods, consequences, and emergency planning, by T.P. McLaughlin, Los Alamos National Laboratory, LA-UR-9l-2325, 12 p., 1991