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Tokaimura Criticality Accident 1999

updated October 2013

- In 1999 three workers received high doses of radiation in a small Japanese plant preparing fuel for an experimental reactor.
- The accident was caused by bringing together too much uranium enriched to a relatively high level, causing a "criticality" (a limited uncontrolled nuclear chain reaction), which continued intermittently for 20 hours.
- A total of 119 people received a radiation dose over 1 mSv from the accident, but only the three operators' doses were above permissible limits. Two of the doses proved fatal.
- The cause of the accident was "human error and serious breaches of safety principles", according to IAEA.

Safety in the nuclear fuel cycle has always been focused on <u>reactor operations</u>, where a huge amount of energy is released continuously in a small volume of material, and where there are substantial amounts of radioactive materials which would be very hazardous if released to the biosphere. A secondary focus is then the <u>high-level wastes</u> from the reactor, which comprise all the potentially hazardous materials from the reactor core.

Other parts of the nuclear fuel cycle have much less potential for widespread harm to people or the environment. They are correspondingly less regulated in some countries, such as Japan.

The Tokaimura plant

The 1999 Tokai-mura accident was in a very small fuel preparation plant operated by Japan Nuclear Fuel Conversion Co. (JCO), a subsidiary of Sumitomo Metal Mining Co. It was not part of the electricity production fuel cycle, nor was it a routine manufacturing operation where operators might be assumed to know their jobs reasonably well.

The particular JCO plant at Tokai was commissioned in 1988 and processed up to 3 tonnes per year of uranium enriched up to 20% U-235, a much higher than for ordinary power reactors. The plant supplied various specialised research and experimental reactors. It uses a wet process.

The approved nuclear fuel preparation procedure involved dissolving uranium oxide (U_3O_8) powder in nitric acid in a dissolution tank, then its transfer as pure uranyl nitrate solution to a storage column for mixing, followed by transfer to a precipitation tank. This tank is surrounded by a water cooling jacket to remove excess heat generated by the exothermic chemical reaction. The prevention of criticality was based upon the general licensing requirements for mass and volume limitation, as well as upon the design of the process. A key part of the design was the storage column with a criticality-safe geometry and allowing careful control of the amount of material transferred to the precipitation tank.

However, the company's work procedure was modified three years earlier, without permission from the regulatory authorities, to allow uranium oxide to be dissolved in stainless steel buckets rather than the dissolution tank. It was then modified further by the operators to speed things up by tipping the solution directly into the precipitation tank. The mixing designed to occur in the storage column was instead undertaken by mechanical stirring in the precipitation tank, thus bypassing the criticality controls. Also there was no proper control of the amount tipped into the hundred-litre precipitation tank, and its shape (450 mm diameter and 660 mm high) enhanced the likelihood of criticality within it.

The accident

On 30 September three workers were preparing a small batch of fuel for the JOYO experimental fast breeder reactor, using uranium enriched to 18.8% U-235. It was JCO's first batch of fuel for that reactor in three years, and no proper qualification and training requirements had been established to prepare those workers for the job. They had previously used this procedure many times with much lower-enriched uranium - less than 5%, and had no understanding of the criticality implications of 18.8% enrichment. At around 10:35, when the volume of solution in the precipitation tank reached about 40 litres, containing about 16 kg U, a critical mass was reached.

At the point of criticality, the nuclear fission chain reaction became self-sustaining and began to emit intense gamma and neutron radiation, triggering alarms. There was no explosion, though fission products were progressively released inside the building. The significance of it being a wet process was that the water in the solution provided neutron moderation, expediting the reaction. (Most fuel preparation plants use dry processes.)

The criticality continued intermittently for about 20 hours. It appears that as the solution boiled vigorously, voids formed and criticality ceased, but as it cooled and voids disappeared, the reaction resumed. The reaction was stopped when cooling water surrounding the precipitation tank was drained away, since this water provided a neutron reflector. Boric acid solution (neutron absorber) was finally was added to the tank to ensure that the contents remained subcritical. These operations exposed 27 workers to some radioactivity. The next task was to install shielding to protect people outside the building from gamma radiation from the fission products in the tank. Neutron radiation had ceased.

The radiation (neutron and gamma) emanated almost entirely from the tank, not from any dispersed materials. Buildings housing nuclear processing facilities such as this are normally maintained at a lower pressure than atmosphere so that air leakage is inward, and any contamination is removed by air filters connected to an exhaust stack. In this case particulate radionuclides generated within the conversion building were collected by the high-efficiency particulate air filters, though noble gases passed through the filters. A smoke test on 5 October confirmed that the negative pressure had been maintained (ie the structural integrity of the building was satisfactory) and that the ventilation system was working. However, owing to the

detection of low levels of iodine-131 being released to the environment through the exhaust, it was later decided to stop ventilation and to rely on the passive confinement provided by the building.

Five hours after the start of the criticality, evacuation commenced of some 161 people from 39 households within a 350 metre radius from the conversion building. They were allowed home two days later after sandbags and other shielding ensured no hazard from residual gamma radiation. Twelve hours after the start of the incident residents within 10 km were asked to stay indoors as a precautionary measure, and this restriction was lifted the following afternoon.



Plan of site from STA, data quoted below from monitoring point A, that from B is slightly lower.

The effects, and analysis

The accident was classified by the Japanese authorities as Level 4 on the International Atomic Energy Agency (IAEA) International Nuclear Event Scale (INES)*, indicating an event without significant off-site risk. It was essentially an 'irradiation' accident, not a 'contamination' accident, as it did not result in any significant release of radioactive materials. Japan's Science & Technology Agency estimated that 2.5 x 1018 fissions had occurred, about half in the first few minutes, releasing 81 MJ (the energy in 2.5 litres of gasoline).

* see table below.

The three workers concerned were hospitalised, two in a critical condition. One died 12 weeks later, another 7 months later. The three had apparently received full-body radiation doses of 16-20,000, 6-10,000 and 1-5000 millisieverts (about 8000 mSv is normally a fatal dose), mainly from neutrons. Another 24 JCO workers received up to 48 mSv. Doses for 436 people were evaluated, 140 based on measurement and 296 on estimated values. None exceeded 50 mSv (the maximum allowable annual dose), though 56 plant workers exposed accidentally ranged up to 23 mSv and a further 21 workers received elevated doses when draining the precipitation tank. Seven workers immediately outside the plant received doses estimated at 6 - 15 mSv (combined neutron and gamma effects). For members of the public, estimates are that one received 24 mSv, four 10-15 mSv, and 15 received 5-10 mSv.

The peak radiation level 90 metres away just outside the nearest site boundary was 0.84 mSv/hr of gamma radiation, but no neutron levels were measured at that stage. The gamma reading then dropped to about half that level after nine hours at which stage 4.5 mSv/hr of neutron radiation was measured there, falling to about 3 mSv/hr after a further two hours, and then both readings falling to zero (or background for gamma) at 20 hours from the start of the criticality.

Neutron dose rates within one kilometre are assumed to be up to ten times the measured gamma rates. Based on activation products in coins from houses near the plant boundary and about 100 m from the reaction, it was estimated that some 100 mSv of neutron radiation would have been received by any occupants over the full period of the criticality. However, the evacuation of everyone within 350 metres of the plant had been ordered 5 hours after the start of the accident. The final report on the accident said that the maximum measured dose to the general public (including local residents) was 16 mSv, and the maximum estimated dose 21 mSv.

While 160 TBq of noble gases and 2 TBq of gaseous iodine were apparently released, little escaped from the building itself. After the criticality had been terminated and shielding was emplaced, radiation levels beyond the JCO site returned to normal.

Only trace levels of radionuclides were detected in the area soon after the accident, and these were short-lived ones. Products from the area would have been as normal, and entirely safe throughout. Radiation levels measured by the IAEA team in residential areas in mid October were at the normal background levels. Measurement of I-131 in soils and vegetation outside the plant showed them to be well under levels of concern for food.

According to the IAEA, the accident "seems to have resulted primarily from human error and serious breaches of safety principles, which together led to a criticality event". The company conceded that it violated both normal safety standards and legal requirements, and criminal charges were laid. The fact that the plant is a boutique operation outside the mainstream nuclear fuel cycle evidently reduced the level of scrutiny it attracted. The state regulator had visited the plant only twice per year, and never when it was operating.

Japan's atomic energy insurance pool said would make a payment to JCO in respect to the accident, its first such payment ever. However, this would be limited to one billion yen, with further liability (the total estimated at 13 billion yen - A\$ 200 million), being met by JCO or its parent company. The plant's operating licence was revoked early in 2000.

Mainstream fuel fabrication plants in Japan are fully automated, engineered to ensure that criticality does not occur, equipped with neutron monitoring systems and fully prepared for any possible criticality accident. Most plants use a dry process in any case, which is intrinsically safer. No major civil reactor uses uranium enriched beyond 5% U-235.

Previous criticality accidents

While this was Japan's first such accident, similar criticality incidents have occurred, especially in US and Russian military plants and laboratories. All but two of these were prior to the early 1980s. Three (in 1958 and 1964) were very similar to this accident. The last of these was the single previous criticality accident at a commercial fuel plant, in USA, resulting in one death.

Of all the previous accidents, 37 occurred in connection with research reactors or laboratory work for military projects, resulting in ten deaths. Another 22 occurred in fuel cycle facilities, all but one military-related, and resulting in seven deaths. The energy released in each of these accidents ranged from about 0.03 MJ to 3 GJ*. The energy released in the similar US accident was about 3 MJ, though due to the prolonged criticality here, some 80 MJ was released, equivalent to the combustion of just over two litres of petrol/gasoline.

* on basis of IPSN report quoting fissions ranging from 10^{15} to 1.2×10^{20} , and each fission yielding 3×10^{-11} Joules. Petrol @ 34 MJ/litre.

The fuel preparation accidents were all in wet processes, due to putting too much uranium-bearing solution in one tank. Mostly these then erupt rather like a saucepan of milk boiling over, and the fission reaction ceases as the material is ejected and dispersed in the immediate vicinity. None of the previous accidents resulted in significant release of radioactivity outside the plants. Practically all were in Russian or US plants, and in reviewing these accidents recently the need for a high level of staff training was emphasised.

The International Nuclear Event Scale

For prompt communication of safety significance

Level, Descriptor Off-Site Impact

Major Accident

On-Site Impact

Defence-in-Depth Degradation

Examples

Chernobyl, Ukraine, 1986 (fuel meltdown and fire); Fukushima Daiichi 1-3, 2011 (fuel damage, radiation release, and evacuation) Mayak at Ozersk, Russia, 1957

6 Significant Release: Full

Major Release:

Widespread health and

environmental effects

For prompt communication of safety significance

Level, Descriptor	Off-Site Impact	On-Site Impact	Defence-in-Depth Degradation	Examples
Serious Accident	implementation of local emergency plans			'Kyshtym' (military reprocessing plant criticality)
5 Accident with Off-Site Risks	<i>Limited Release:</i> Partial implementation of local emergency plans	Severe core damage		Three Mile Island, USA, 1979 (fuel melting). Windscale, UK, 1957 (military).
4 Accident Mainly in Installation either of:	<i>Minor Release:</i> Public exposure of the order of prescribed limits	Partial core damage. Acute health effects to workers		Saint-Laurent A1, France, 1969 (fuel rupture) & A2, 1980 (graphite overheating) Tokai-mura, Japan, Sept 1999 (criticality in fuel plant for an experimental reactor).
3 Serious Incident any of:	<i>Very Small Release:</i> Public exposure at a fraction of prescribed limits	Major contamination, Overexposure of workers	Near Accident. Loss of Defence-in-Depth provisions	Fukushima Daiichi 4, 2011 (fuel pond overheating); Fukushima Daini 1, 2, 4, 2011 (interruption to cooling); Vandellos, Spain, 1989 (turbine fire); Davis-Besse, USA, 2002 (severe corrosion); Paks, Hungary 2003 (fuel damage
2 Incident	nil	nil	Incidents with potential safety consequences	
1 Anomaly	nil	nil	Deviations from authorised functional domains	
0 Below Scale	nil	nil	No safety significance	

Source: International Atomic Energy Agency

Sources:

ENS NucNet news # 397-402, 409, 410, 414 & 459/99, 36/00, 169/00 background # 10-12/99. <u>IAEA</u> Report on the Preliminary fact-finding mission, IPSN 1/10/99, Yomiuri Shimbun 4/11/99 Atoms in Japan, Dec 1999

A Review of Criticality Accidents, 2000 Revision, DOE Los Alamos laboratory, LA-13638.