2. G. E. HANSEN, "Assembly of Fissionable Material in the Presence of a Weak Neutron Source," *Nucl. Sci. Eng.*, 8, 709 (1960).

3. D. L. HETRICK, *Dynamics of Nuclear Reactors*, University of Chicago Press, Chicago (1971).

5. Criticality Incident—August 24, 1970, Windscale Works, *J. T. Daniels (UKAEA-Risley), H. Howells, T. G. Hughes (UKAEA-Windscale)*

At 18.15 on August 24, 1970, the criticality alarms sounded[1] in the Plutonium Recovery Plant, building B.203 and in nearby building B.277, at the Windscale Works of the United Kingdom Atomic Energy Authority, Production Group.

The two buildings were evacuated promptly and the staff assembled in the Criticality Control Center. It was quickly established that building B.277 contained no fissile material at the time and could be discounted as the source of radiation. The two men who had been working in the B.203 plant were monitored immediately on arrival at the Criticality Control Center, and this showed no personal contamination and no indication of significant irradiation. Examination of dosimeters and measurement of ^{24}Na in a whole-body monitor confirmed that neutron doses, if any, were very low. These preliminary measurements were completed by 23.45 on the same day. More extensive whole-body monitoring and subsequent investigations on August 25 confirmed that the whole-body doses received by the two men were <1 and 2 rads, respectively.

Reentry surveys by Health Physics staff commenced at 18.25 on August 24 and by 18.35 radiation levels had been confirmed as normal in building B.277. However, abnormal levels up to 200 mR/h γ had been detected in B.203 at the main control area on the third floor and 40 mR/h γ at the vessel ventilation extract filter on the second floor. Subsequent monitoring showed these levels to be decaying rapidly, indicating the presence of short-lived fission products, thus confirming that a criticality incident had occurred despite the absence of significant radiation exposure. A small release (~5 mCi) of gaseous and particulate fission products occurred via the vessel extract filters and stack (400 ft high), but the effects were not detectable at ground level.

The B.203 plant was designed as the Plutonium Purification Section of the original Irradiated Fuel Reprocessing Plant. In 1964 it was adapted to recover plutonium by the TBP/OK solvent extraction process from both solid and liquid residues. These residues include oxides, mixed Pu/U oxides, fluorides, nitrates, slag, and oxalate mother liquors. In addition to the residues, aqueous raffinates, solvent wash liquors, and off-specification product have been recycled from time to time. The plant consists of two parallel lines, North and South, housed in cells with walls ~12 in. thick. Extraction from nitrate solution is carried out in 3 pulsed and 1 static column, all of which are geometrically safe. The extraction columns are preceded by dissolver units, conditioners, and constant volume feeders (CVF), all of which are either mass or concentration limited for criticality control. Solutions are transferred from dissolvers to conditioners and conditioners to CVFs by a vacuum lift system via closed transfer vessels. The outlet of the transfer vessels to the CVFs is by a bottom drain, $\frac{1}{2}$-in. bore, having a 25-ft-deep lute. The 2-ft-diam × 2-ft, 3-in.-high transfer vessel has convex dished ends.

A recovery program commenced on August 14 and on August 24 oxide material was being processed in the North Cell. The incident occurred as a lift of solution from a conditioner to a vacuum transfer vessel was ending. At this time the transfer vessel contained the maximum amount of solution, as the draining part of the cycle does not commence until the vacuum breaks; i.e., when the conditioning vessel has been emptied.

The excursion was small and there was insufficient irradiation of surrounding materials to permit an estimate of the yield by activation analysis. However, measurements of fission product decay rates in the vicinity and spectrometric analysis of fission products in the aqueous solution led to the conclusion that the total yield was of the order of 10^{15} fissions. Consideration of the response of the alarm systems to an incident of this size led to the conclusion that the duration of criticality was probably from 5 to 10 sec.

Post-incident investigations were conducted with great caution, due to the following:

1. Aqueous solution subsequently syphoned from the CVF contained fission products, but at a plutonium concentration of 6 g/liter this solution could not have been the main source of reactivity.

2. The yield of 10^{15} fissions was too small to have provided an inherent shutdown mechanism; e.g., boiling.

It was decided, therefore, that the bulk of the fissile material that had been critical was still within the transfer vessel and that it was likely it was only just subcritical. It was also believed that as such a small yield could only result from a narrow band of conditions, it was likely there had been some mechanism that had added reactivity incrementally. In these circumstances any small disturbance or increase of reflection could have resulted in another, possibly bigger, excursion.

Subsequent remote investigations, which include gamma and neutron monitoring, gamma radiography, intrascope photography, and location of a liquor surface with a conductivity probe, established that even though the transfer vessel should have been empty it still contained liquor to a depth of $8\frac{1}{2}$ in. associated with kilogram quantities of plutonium. This solution was removed remotely in $2\frac{1}{2}$-liter aliquots by a suction pipe fitted with a conductivity probe inserted from above through the $\frac{1}{2}$-in. bore vacuum line. Approximately 40 liters of liquor was removed from the vessel and analysis showed it to be solvent with a plutonium concentration of 55 g/liter. Thus, at the time of criticality, the contents of the transfer vessel were 40 liters of 20% TBP/OK solvent having 55 g/liter plutonium in solution, together with 50 liters of 7 M HNO3 having 6 to 7 g/liter plutonium in solution. There may have been a small quantity of solids containing plutonium in the vessel but it would not have added significantly to the reactivity. The two phases, solvent and aqueous, were largely separated, the density of solvent phase being 0.96 g/ml and that of the aqueous phase 1.3 g/ml.

The source of the solvent has not been identified positively but there are several operations in which aqueous solutions containing both entrained and dissolved solvent could have been fed into the conditioners. Calculations based on solvent degradation products indicate that the solvent was almost certainly in the transfer-vessel lute system before the August 1970 campaign began, and may have accumulated over a period of one or two years.

Trials conducted on an inactive transparent replica of the system demonstrated that, if solvent is present, as aqueous solution is lifted into the transfer vessel the phases tend to separate out while the lift is in progress, and the aqueous solution forms a layer at the bottom of the transfer vessel. When the vacuum breaks, allowing the contents to drain, only the aqueous liquor drains via the lute to the CVF and the solvent remains trapped as a supernatant layer. The practice of washing out the plant periodically with acid would not have displaced the solvent from the lute but may have reduced its plutonium concentration.

A straightforward mechanism can be postulated for the buildup of plutonium once solvent is trapped in the transfer vessel. Successive batches of conditioned plutonium nitrate at 7 g/liter of plutonium would come into contact with solvent, and plutonium would be extracted into the solvent each time a transfer took place via the transfer vessel. A study of the buildup of plutonium in solvent suggests that at the plutonium concentrations and acidities that existed at the time of criticality, the last cycle probably involved a plutonium transfer from aqueous to solvent as low as 30 g, thus providing a small incremental increase of reactivity. The energy released from as few as 10^{15} fissions spread over several seconds would not have been sufficient to provide a shutdown mechanism due to the phenomena observed in other criticality incidents; viz., boiling, physical expulsion of solution, or microbubble formation. We are led to the conclusion, therefore, that the contents of the vessel passed through transient geometric configurations that involved the increase of reactivity, which produced criticality and also contained a mechanism that shut the reaction down. Observation of the replica system showed that as the aqueous phase flowed into the transfer vessel it fell as a streamlined jet into the solvent layer. In doing so there was some dispersion that generated an interface band of emulsion consisting mainly of globules of solvent in the aqueous phase. This emulsion band was ~5 cm thick below the jet. Calculations[2] using the MONK Monte Carlo program show that the presence of the jet of aqueous solution, having a plutonium concentration of <7 g/liter, created a "hole" in the middle of the solvent layer and so decreased its reactivity as long as the flow continued. On the other hand, the presence of the emulsion band produced a more reactive system than that which existed in the quiescent state with the phases separated after flow had ceased. Laboratory tests with the solvent removed from the transfer vessel after the incident led to an estimate of between 5 and 10 sec for the collapse of the emulsion band after the cessation of flow. Thus, it seems likely that at the cessation of flow the system became prompt critical and the shutdown mechanism was the collapse of the interface emulsion layer. The separated quiescent phases were shown to be just subcritical.

The post-incident investigation showed there had been no breach of the criticality clearance conditions. The Board of Enquiry recommended, *inter alia*, that means of detecting and removing plutonium accumulations should be provided in all vessels in the plant, other than those that are safe by shape.

The plant has now been recommissioned with neutron monitors installed on all vessels that are not safe by shape. The vessel drain lutes have also been modified to facilitate positive drainage and washout procedures.

1. J. T. DANIELS, H. HOWELLS, and T. G. HUGHES, "Criticality Incident in the Plutonium Recovery Plant at the Windscale Works of the United Kingdom Atomic Energy Authority," to be published by the UKAEA (1971).

2. J. H. CHALMERS, "Criticality Calculations Relating to an Incident at Windscale," AHSB(S)R.195, UKAEA (1971).

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"FRENCH EXCURSION EXPERIMENTS"