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résumé

Quelle est la quantité de plutonium qui sera produite dans les réacteurs de puissance thermiques et quelle sera sa valeur? Le coût de fabrication des assemblages combustibles au plutonium pour le recyclage dans les réacteurs thermiques donnera-t-il au plutonium une valeur effectivement négative? Le coût de stockage du plutonium justifie-t-il sa plus grande valeur éventuelle dans les réacteurs surrégénérateurs rapides? Ces questions intéressent actuellement beaucoup les entreprises de service public utilisant des centrales nucléaires, surtout aux Etats-Unis où la fin de l'accord de rachat de l'AEC a fait ressortir les problèmes associés au plutonium. Les fabricants de combustible dans le monde entier s'efforcent de créer des matériels de fabrication de combustible au plutonium qui seront commercialement économiques malgré les difficultés que présente le traitement du matériau le plus toxique qui soit connu. Cette situation intéressante est examinée de près dans un article spécial dans ce numéro. Un autre article intéressant, rédigé par T. G. Hughes, décrit un incident de criticalité qui s'est produit dans une usine de récupération de déchets de plutonium à Windscale. Quoique l'incident était d'importance minime, sans occasionner de dégât, il sert à illustrer le besoin d'examiner les mécanismes d'accumulation les plus imprévus dans les installations de traitement de matériaux fissiles.

Dans son article, R. M. Hogg décrit une nouvelle méthode de traitement des déchets gazeux provenant de centrales nucléaires par absorption liquide-gaz, qui a été développée par Babcock & Wilcox aux Etats-Unis. Un appareillage d'essais, permettant de mesurer le fluage de compression dans les matériaux combustibles à céramique en cours d'irradiation, a été employé à Harwell ainsi que pour des études sur le bioxyde d'uranium. Le développement et les performances de cet appareillage sont décrits dans un article de R. W. Stratton.

kurzreferate

Wieviel Plutonium wird in den thermischen Leistungsreaktoren der Welt erzeugt werden, und was wird sein Wert sein? Werden die Kosten der Herstellung von Plutonium-Brennelementen für thermische Reaktoren dem Plutonium einen effektiv negativen Wert geben? Werden die Kosten der Lagerung von Plutonium seinen späteren höheren Wert in schnellen Brutreaktoren rechtfertigen? Diese Fragen sind gegenwärtig von grosser Bedeutung für Betreiber von Kernkraftwerken, besonders in den USA, wo das Aufhören des Rückkaufs durch die AEC der Frage, was mit dem Plutonium geschehen soll, ganz besondere Dringlichkeit gegeben hat. Brennstoffhersteller in aller Welt bemühen sich sehr um die Erstellung von Fertigungseinrichtungen, die trotz der Schwierigkeiten bei der Hantierung des giftigsten aller bekannten Stoffe kommerziell wirtschaftlich sind. Diese interessante Situation wird in dieser Ausgabe in einem Sonderbericht eingehend untersucht.

Ein aufschlussreicher Artikel von T. G. Hughes beschreibt einen Kritikalitätsunfall in einer Plutoniumschrott-Aufarbeitungsanlage in Windscale. Obgleich es sich um einen äusserst kleinen Unfall handelte, der keinen Schaden verursachte, wird dadurch nachdrücklich auf die Notwendigkeit hingewiesen, auch ganz unerwartete Mechanismen der Bildung kritischer Massen in Anlagen zu untersuchen, in denen Spaltstoffe hantiert werden.

Ein neues Verfahren zur Hantierung von gasförmigen Abfällen von Kernkraftwerken durch Flüssigkeits-Gasabsorption wurde von Babcock & Wilcox in den USA entwickelt; es wird in einem Artikel von R. M. Hogg beschrieben.

Ein Testrig, das Messung des Kriechens unter Druckspannung in keramischen Brennstoffen bei Bestrahlung gestattet, wurde in Harwell zu Untersuchungen an Urandioxid benutzt. Die Entwicklung und das Betriebsverhalten dieses Rigs werden in einem Artikel von R. W. Stratton beschrieben.

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Front Cover

The PFR fuel line at Windscale—the equipment in glove boxes is controlled from behind a further sealed face.

Criticality incident at Windscale

By T. G. Hughes, *BNFL, Windscale*

Since the inception of the nuclear industry in 1942, some twelve criticality incidents associated with reprocessing, recovery or fuel plants have been reported throughout the world. This comparatively small number has been due to the extreme care which has been taken in both the design of the nuclear plants and the detailed operational procedures that have been adopted. In Britain there has been only one such incident since the start of the industry some twenty years ago. This occurred on the evening of August 24, 1970.

In the nuclear industry a criticality incident may be described as an unsolicited nuclear excursion brought about by accidental accumulation of a supercritical quantity of fissile material. The possibility of such an incident is always present in the reprocessing of irradiated fuel, the manufacture of certain types of nuclear fuel, and also fissile material recovery associated with both these processes. All three of these processes are undertaken at the Windscale factory of British Nuclear Fuels Limited (BNFL) and in each case the daily throughput of fissile material can amount to several tens of kilograms.

At Windscale an extensive system of very sensitive radiation monitoring instruments connected to an audible alarm mechanism is provided in all plants where a criticality excursion may occur. The disposition of the instruments is such that an immediate alarm would be given if a criticality excursion occurred in any conceivable area of the plant. Associated with the system is a precisely defined and well drilled personnel evacuation procedure. On the evening of 24 August 1970 this alarm system was activated in two adjacent buildings. One of them was a new production plant for the manufacture of plutonium-containing fuel for the British prototype fast reactor. The other was a plant used to recover plutonium from miscellaneous residues. The two buildings were evacuated promptly and the staff assembled in the criticality control centre. The control centre is the focal point of all the criticality instrumentation of the chemical processing area and is equipped to deal with personnel problems resulting from an actual incident. Examination of personal dosimeters of individuals from both buildings gave no indication of any significant radiation uptake.

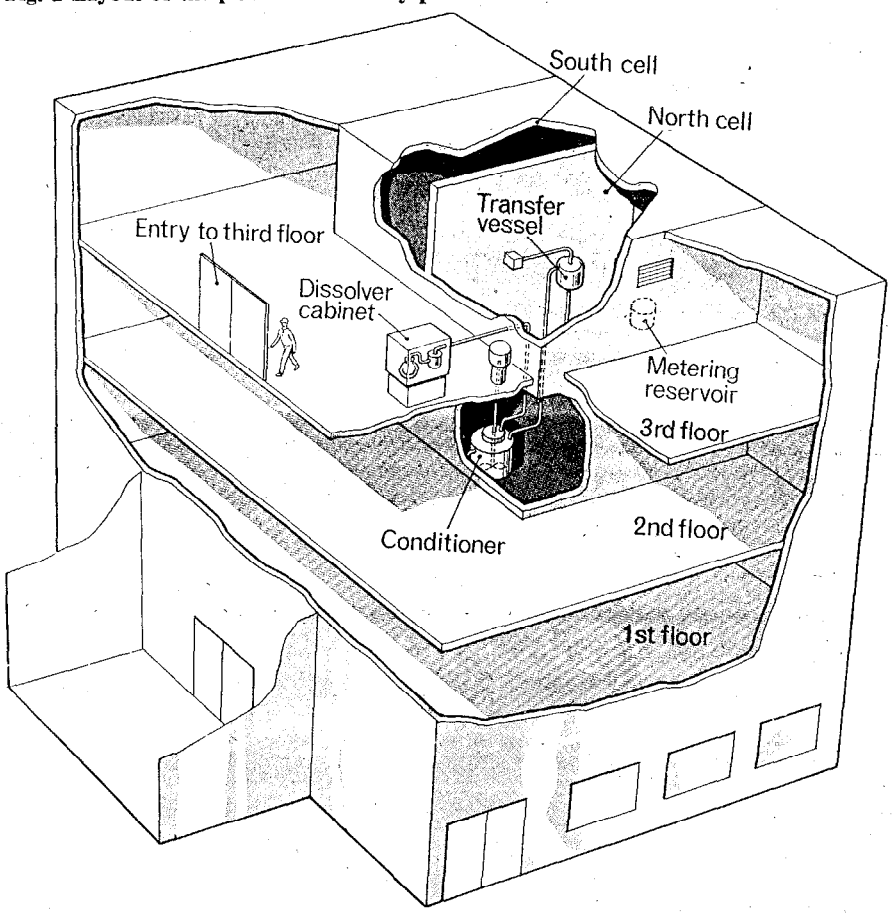
While the personnel checks were being undertaken, re-entry surveys of the two buildings in which the alarms had sounded were started. The plutonium fuel plant was readily eliminated. This plant had just completed inactive commissioning trials and in fact the first charge of plutonium oxide was actually in transit to the building when the alarm sounded. Checks in the plutonium recovery plant showed radiation levels higher than normal but certainly lower than those which would have been predicted following a criticality excursion.

Plutonium recovery plant

The plutonium recovery plant consists of two identical lines of equipment housed in separate cells with a common dividing wall. The cells are effectively concrete boxes with one foot thick concrete walls. Surrounding these walls on three sides is a conventional building shell, housing the control equipment located on four floors. This is shown diagrammatically in Figure 1.

A simplified line diagram of the main plant items of a processing line is shown in Figure 2. A glove box (1) located in the inactive operating area houses a glass vessel (2) in which plutonium residues may be dissolved in nitric acid. The solution produced may be passed via a filter (3) through the wall of the concrete cell to a stirred treatment vessel (4). Liquors from the latter may be lifted by the application of vacuum to a transfer pot (5). When all the liquor from the treatment vessel has been lifted into the transfer pot the vacuum breaks and the liquor may then flow via a lute into the metering reservoir (6).

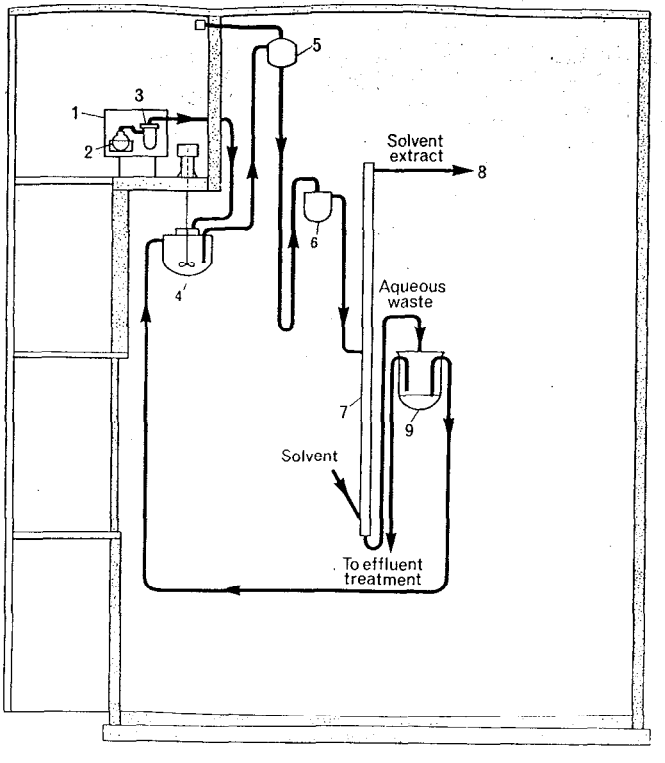
Fig. 1 Layout of the plutonium recovery plant



(Plant items (1) to (5) are in fact duplicated but have been omitted from the diagram in the interests of clarity.) From here, the liquor may be metered to a pulsed column (7) for the first stage of solvent extraction which constitutes the purification process. In the column the plutonium is extracted from the aqueous solution by a solvent —TBP/kerosene—which rises through the column and leaves at point (8). The aqueous solution from which plutonium has been removed flows down the column and via a lute into a waste liquor collecting tank (9). Provision is made to enable liquors to be returned from this tank to the original treatment vessel (2) if for some reason the extraction of plutonium from the aqueous solution has been inadequate. When the incident occurred, only one of the processing lines, the so-called north unit, was in operation. The south unit was nominally clean of plutonium solution having been washed out with inactive reagents at the end of a previous campaign.

The re-entry survey had indicated that the seat of the excursion was in the region of the two treatment vessels and their associated vacuum lift pots. At the time the alarm sounded liquor was being transferred from one of the treatment tanks through the lift pot to the metering reservoir. In addition another batch of plutonium liquor was being treated chemically in the second treatment vessel. A sample of the latter was taken and indicated no abnormality. However, a sample taken from the metering reservoir showed the presence of fission products that would have been produced by a nuclear excursion, although the plutonium concentration of this solution was only 6 gm/litre which was perfectly normal. Complete proof that the liquor in the reservoir was not the main source of fissile material responsible for the incident was established by siphoning the contents of the vessel into containers which were themselves safe by geometry and performing a complete analytical assay. Suspicion was therefore concentrated on the transfer pot (5). This vessel was positioned in the direct line of an air intake louvre in the cell wall through which radiation monitoring instruments could be introduced. The measurements taken confirmed that the incident had occurred in the transfer pot which almost certainly contained the bulk of the fissile material which had been critical.

Measurement of the fission product decay rates in the vicinity of the transfer pot and spectrometric analysis of the fission products in the aqueous solution from the metering reservoir led to the conclusion that the total yield of the excursion was 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 no more than a few seconds. It was argued that such a small excursion could only result from a very narrow band of conditions and it was likely that there had been some mechanism that had added reactivity incrementally.



1 – Plutonium dissolving glove box
2 – Glass dissolver vessel
3 – Plutonium solution filter
4 – Treatment vessel
5 – Transfer pot
6 – Plutonium solution metering reservoir
7 – Pulsed extraction column
8 – Solvent outlet line for column to next stage of process
9 – Aqueous raffinate waste tank

Fig. 2 Flow diagram for the plutonium recovery plant

Finding the cause

In attempting to assess the cause of the incident, the possibility of the presence of solid plutonium compounds seemed the most likely. Such a situation could have arisen if solids remaining at the dissolution stage had bypassed the filter. It was decided to carry out a thorough check of the vessels in the south unit before proceeding in any way to ascertain the precise cause of the incident. This check did indicate the presence of solids in some vessels but the quantities involved were well below the level required to initiate a criticality excursion. Although a sensible precaution, this procedure gave no hint of the eventual reason for the criticality incident, although it greatly assisted in planning monitoring procedures in the affected cell. These included gamma and neutron monitoring, thermal profile measurement and gamma radiography. An assessment of the results of these measurements indicated that the transfer pot did contain appreciable quantities of fissile material and enabled a rough contour of its distribution to be worked out. It was of interest to note that the detailed interpretation of these readings depended on an assumption of the geometry of the fissile material inside the vessel. This led to estimates varying from 1·5 kg to 25 kg plutonium. The very high level of the latter figure, derived from neutron measurements, was entirely due to the assumption that the plutonium was present as a layer of solid and not, as it turned out, in an appreciable volume of homogeneous solution.

It was eventually decided to gain access via a $\frac{3}{4}$in diameter inlet line to the centre of the top of the transfer pot. This line under normal circumstances was connected to an air ejector which produced the transfer vacuum. To do this without entering the cell meant drilling a 6in diameter hole with a diamond drill through the reinforced concrete roof. This was achieved satisfactorily and the pipe was manually cut using a hacksaw.

While the preparatory work to gain access to the vessel was under way, methods of viewing inside the vessel through the $\frac{3}{4}$in hole were being investigated. The photographic department of AWRE at Aldermaston produced a system based on fibre optics which showed great promise. Several instruments were manufactured, one of which gave excellent results. The instrument established that the transfer

pot contained an appreciable quantity of liquor. A conductivity probe was used to establish the precise depth of liquor in the vessel. This indicated a depth of 8½in, equivalent to a volume of approximately 40 litre. On the basis of simple hydraulics the vessel should have been empty and the presence of liquid indicated a blockage at the outlet which could have arisen from an accumulation of solids in this area.

It was decided to empty the liquor from the transfer pot in 2½ litre aliquots using flexible plastic piping. This was achieved satisfactorily, the transfer being effected by siphoning to a collection point in an adjacent building. The first aliquot produced the complete explanation for the incident. The liquor was found to be a solution of plutonium nitrate in TBP/kerosene solvent. The solution had a specific gravity of 0·96 gm/ml and contained 55 gm plutonium/litre. It was now clear that at the point of criticality the vessel would have contained some 2½ kg plutonium in the combined 40 litre of solvent and the 50 litre of aqueous solution which was being transferred. The specific gravity of the aqueous liquor was 1·3 gm/ml, which, considering the geometry of the lute system, was high enough to ensure that the solvent, once having entered the transfer vessel, was locked there permanently. Subsequent examination of the empty vessel using the fibre optic equipment showed that the vessel contained virtually no solids and the outlet was not blocked.

Source of the solvent

The source of the solvent has not been positively identified. Recycled aqueous raffinate from the extraction column is one possibility. In this way solvent could have been transferred by simple entrainment, the latter being enhanced under conditions of incipient emulsification which incidentally could have been the cause of poor plutonium extraction in the pulsed column, leading to plutonium levels in the raffinate which justified recovery by recycle. Detailed chemical examination of the solvent indicated a high degree of degradation which had been caused by the alpha irradiation associated with the presence of plutonium. The precise age of the solvent was difficult to define. Various estimates, between several months and over two years can be made, depending on the estimated rate of accumulation. The plant had been in operation since 1954 without any previous indication of such an accumulation.

Trials were carried out using a transparent replica of the treatment vessel/transfer pot/metering reservoir system. These showed that under the conditions prevailing at the time of the incident the aqueous solution lifted into the transfer pot from the treatment vessel rapidly separated from the solvent already present in the pot. This condition existed until transfer of the aqueous solution had been completed and the vacuum had broken. At this point there was fairly violent agitation, but even so, no intimate mixing of the two phases.

A straightforward mechanism can be postulated for the build up of plutonium once solvent is trapped in the transfer vessel. At the acidity of a normally treated batch of aqueous liquor the partition coefficient of plutonium between the aqueous and solvent phases is heavily in favour of the transfer of plutonium to a solvent phase. Hence, as successive batches of plutonium nitrate aqueous solution pass into and through the transfer pot, plutonium progressively builds up in the trapped solvent phase. This applies despite the relatively poor mixing that occurs during transfer. As the concentration of plutonium in the solvent phase increases the rate of transfer from the aqueous phase would reduce.

Operation of excursion

A detailed assessment of the operation indicated that on the occasion of the criticality excursion a transfer of a few 10's of grams of plutonium would probably have occurred from the aqueous to the solvent phase. This could have provided the small incremental increase in activity giving rise to the excursion. The energy released from as few as 10^{15} fissions spread over a couple of seconds would not have been sufficient to provide a shutdown mechanism due to the phenomena observed in other criticality incidents, for example, boiling, physical expulsion of solution or microbubble formation. It therefore seems highly likely that the contents of the vessel passed through transient geometric configurations which involved the increase of reactivity which produced criticality, and also contained the mechanism which shut the reaction down.

Observation of the replica system showed that as the aqueous phase flowed into the transfer vessel it poured as a streamlined jet into the solvent layer. In doing so there was some dispersion which generated an interface band of emulsion some 5 cm thick consisting mainly of globules of solvent in the aqueous phase. Computer calculations using the well established MONK Monte Carlo programme (an adaptation of the original GEM code) showed that the presence of the jet of aqueous solution, having a plutonium concentration of less than 7 gm/litre, 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 about five seconds for the emulsion band to collapse 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 calculated by the computer programme to be just subcritical.

Board of Enquiry

Immediately following the incident a Board of Enquiry was set up consisting of experts from throughout the U.K. Atomic Energy Authority. The measures taken on the plant to establish the cause of the incident and to render the plant safe were endorsed by the Board. A report was produced making firm recommendations for plant modifications to prevent a recurrence of such an incident. The two most important of these were that neutron monitors should be installed on all vessels which are not safe by shape and that provision be made to enable luted plant items to be emptied completely.

The time scale from the moment of the incident to the emptying of the transfer pot, and eventually the rehabilitation of the plant, was probably longer than the above brief account might suggest to the reader. In fact the work leading up to the emptying of the transfer pot took nearly five weeks. Subsequently a further ten weeks were required to effect modifications recommended by the Board of Enquiry. Normal operations of the plutonium recovery plant recommenced on 6 December 1970 and have proceeded smoothly since that date.

The incident differed significantly from any previously recorded throughout the world. There was no significant uptake of radiation to any individual and no spread of contamination outside the active plant containment. All other plutonium processing units have been examined in detail to ensure that they conform in all respects with the recommendations of the Board of Enquiry which have become mandatory for all future plant design.