

Commissariat à l'Energie Atomique, France.

Light-water reactors have a tritium balance which should be considered from both the working constraint and environmental pollution aspects.

Formation in the primary circuit :

In the two categories of light-water nuclear stations, the pressurized reactor, which uses boric acid in solution for reactivity control, is conspicuous for the amounts of tritium formed in the primary circuit. If both deuterium activation in the water, and fission tritium diffusion in the case of zircaloy-clad fuel are negligible sources compared with the nuclear transmutation of boron-10, a model proposed by LANGECKER and GRAUPE [1], (checked by the tritium balance of the RHEINSBERG reactor) can be used to estimate the tritium production in a hypothetical 1000 MWe pressurized reactor using boric acid for chemical reactivity control [2].

The leakage of the circuit limits the growth of the tritium activity, which thus passes through a maximum before the end of the annual running cycle rather than reaching an equilibrium concentration just as the cycle ends, Figure 1 shows the influence of the leakage rate on the tritium concentration maximum in the primary circuit of the reactor working at nominal power [2].

The rise time to maximum concentration is also dependent on the leakage rate. However, in the hypothetical 1000 MWe reactor, the dilution of the primary circuit water when the vessel is opened, at the time of fuel changes, can quickly raise the tritium concentration in the pool so much that work is harmful in the presence of such a large enclosed free surface of water, unless suitable precautions are taken. The simplest means of slowing down the tritium contamination of pool water is by a controlled discharge, its time and place in the annual running cycle depending on the leakage rate.

The long-term variation in the tritium concentration of the primary circuit water, and of the pool water after dilution, is estimated as a function of the leakage rate and of various types of discharge. Figure 2 gives as an example the development of the tritium activities in the primary circuit water and in the pool against time, for a leakage rate of 0.0012 d^{-1} and various types of primary water discharge [2].

* Paper to be presented at the 1976 Reactor Meeting of Deutsches Atomforum/Kerntechnische Gesellschaft, 30 March - 2 April DUSSELDORF.

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Reactor congress. Dusseldorf (F.R. Germany), 30 March
2 April 1976

CEA-CONF--3478

FR7602577

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The total annual production of tritium in the primary circuit water is obviously dependent on the variation of the boric acid concentration. For the hypothetical reactor in question, working as a base load station, a linear decrease from 1500 ppm natural boron at the start of the annual cycle to about 400 ppm at the end was assumed. The annual tritium production under these conditions is about 530 Ci. All estimations concerning this reactor neglect the natural disappearance of tritium by radioactive decay. The peak tritium concentration in the primary circuit attains approximately $2 \cdot 10^{-3} \text{ Ci.l}^{-1}$ during the first year at power. It develops then slowly, reaching an equilibrium value of the order of $2,5 \cdot 10^{-3} \text{ Ci.l}^{-1}$ after several years. Its maximum concentration at the beginning of a controlled discharge operation can be of this order.

Production in the fuel :

The tritium produced by ternary fission in the fuel is much more abundant and remains almost entirely confined in the uranium oxide if the fuel is clad with zircaloy. The quantity formed annually is more than 20 times larger than that produced in the primary circuit water, and reaches about 12,400 Ci. With a certain time lag and allowing for radioactive decay this tritium is all sent into the effluent of a fuel reprocessing plant.

Elimination of tritium :

Attempts have been made recently to forecast environmental pollution by the tritium of electronuclear stations [3] [4], on the basis of the predicted development of different types of reactors in western electronuclear equipment. They illustrate the kind of urgency of detritiation units associated with electronuclear plants.

Since a large reprocessing plant is supposed to be able to treat fuel from 20 to 50 electronuclear stations, the highest priority should be given there to the appropriate confinement of tritium.

The American process known as "voloxidation" consists of roasting the fuel, already cut into pieces and before dissolution, then separating the tritium from the sweeping gaz. This method has certain disadvantages and another process, whereby the tritium is highly concentrated by recycling the dissolution reagent, is contemplated in France [5]. According to present estimates, the result would be a reduced quantity of effluent with a tritium concentration nearly 1000 times higher than that of the primary circuit water in the hypothetical 1000 MWe reactor. If it is then agreed to detritiate the water from spent fuel dissolution in reprocessing plants, the total tritium discharge from pressurized water reactors (by leakage and controlled discharge) becomes so limited that only local environment conditions will need to be taken into account until the end of this century, according to [3] and [4].

Tritium enrichment processes :

The hydrogen isotope separation methods used on a large scale for the production of heavy water should naturally be

considered first, because of present technological experience. There is nevertheless a fundamental difference between an efficient detritiation and an optimised heavy water manufacturing process [6]. Much more elaborate safety systems are also necessary, but the running of a heavy water detritiation unit attached to the Grenoble High-Flux Reactor already provides useful experience.

An economic study carried out at the CEA has dealt with the special case of the detritiation of a daily reprocessing plant output of 1.5 ton water with a tritium activity of $2,3 \text{ Ci l}^{-1}$, the decontamination factor desired being 100. Amongst the processes directly applicable to water, i.e. essentially distillation, chemical exchange $\text{H}_2\text{O}-\text{H}_2\text{S}$ and electrolysis, the choice cannot depend exclusively on the estimated costs per kilogram of water treated. Distillation would appear tempting because of its ease of operation and relative safety, whereas chemical exchange between water and hydrogen sulphide is much cheaper and would be ideal, on the assumption that the need for economy warrants much more stringent plant working conditions. Electrolysis consumes much more energy but could serve as a preliminary transformation stage, in the same way as catalytic exchange between hydrogen gas and tritium or decomposition on suitable contact (which in fact still need serious development work) to produce hydrogen for gaseous isotope separation processes. Of these latter, gas phase chromatography on a fixed palladised phase has distinct technological advantages but suffers the handicap of a large investment in precious metal [7]; chemical exchange between hydrogen and ammonia, in spite of a very high separation factor, has the chief disadvantage of working at very high pressure, while cryogenic distillation of hydrogen costs much the same as water distillation but is subject to coercions in operation.

Therefore various processes may be considered in order to remove tritium from aqueous waste in fuel reprocessing plants. But the development of methods to be applied on tritium removal from reactor water itself is of another order of difficulty owing to the very low concentration (approximately $2.10^{-3} \text{ Ci.l}^{-1}$, corresponding to about $1,25.10^{-9}$ in concentration of HTO molecules), only detailed study of the processes will allow a choice when the time comes.

It is however an interesting result that several solutions may be considered both technically possible and economically tolerable.

References :

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Relative maximum value of tritium activity (Ci.l⁻¹)

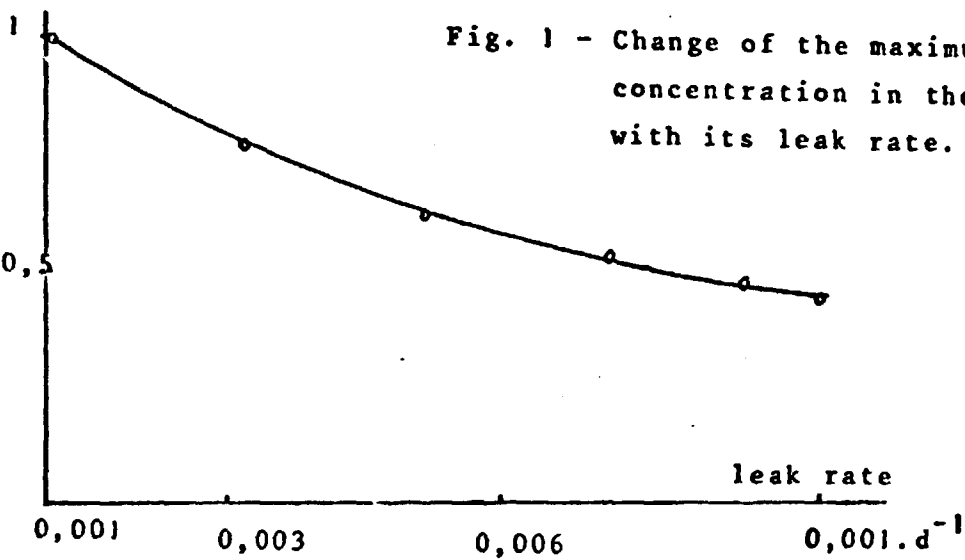


Fig. 1 - Change of the maximum tritium concentration in the primary circuit with its leak rate.

Fig. 2 - Development of tritium activity in the water of a pressurized 1000 MWe reactor with boric acid reactivity control.

Leak rate $F = 0,0012.d^{-1}$

Replacement of the cooling water in the pressure vessel and one primary loop before refueling
(other primary loops being isolated) _____ primary circuit

Extraction of cooling water equivalent to the volume of the pressure vessel and one primary loop before refueling ----- primary circuit
..... pool

Controlled discharge of 1.5 times the volume of the primary circuit at maximum tritium concentration
— : — : primary circuit
- - - - - pool

