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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

**SENA SAFETY EVALUATION**

**FACTORS INVOLVED IN THE EVALUATION OF THE MAXIMUM  
CREDIBLE BORON RELEASE FROM THE CORE  
SURFACES OF A PWR WITH CHEMICAL SHIM**

by

**G. PETRANGELI  
(CNEN)**

1967



Report prepared by the CNEN  
Comitato Nazionale per l'Energia Nucleare, Rome - Italy

Safeguards and Controls

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Experimental data on the phenomena possibly leading to boron deposition and release from core surfaces have been collected from the open literature, critically evaluated and presented in this report. A method is also described for the evaluation of an upper limit of the accidental reactivity insertion.

The results of the application of this method to practical cases concerning pressurized water reactors of current design indicate that an upper limit for the reactivity insertion is of the order of a few tenths of a percent in  $\Delta k/k$ .

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### SUMMARY

The purpose of the present report is to offer a basis for the evaluation of an upper limit of reactivity input for the commonly named "boron-hideout accident". This accident would be caused by the sudden removal from core surfaces of previously deposited boron.

The following items are briefly treated: phenomena leading to boron deposition on core surfaces; evaluation of the amount of crud deposited on core surfaces; evaluation of the amount of boron absorbed by crud; boron deposition due to the presence of alkali metaborates; boron depositions due to boric acid concentration on heat transfer surfaces; percentage of deposited crud which can be released during thermal transients; evaluation of reactivity release in a "boron-hideout accident".

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# FACTORS INVOLVED IN THE EVALUATION OF THE MAXIMUM CREDIBLE BORON RELEASE FROM THE CORE SURFACES OF A PWR WITH CHEMICAL SHIM (\*)

## 1 — INTRODUCTION

The use of dissolved boron in the primary water of pressurized water reactors in order to control long-term reactivity effects faces the safety analyst with the problem of evaluating the possibility of occurrence and the consequences of accidental situations arising from this mode of operation.

The postulated accident consists in the rapid removal from core surfaces of previously deposited boron as a consequence of thermal or hydraulic transients, and in the consequent power excursion.

The experience so far available with PWR's using chemical shim amounts to several reactor-years of operation. A large part of this experience comes from the operation of the Saxton reactor under close reactivity control in order to detect even small reactivity effects; the precision of this reactivity control has been probably such to allow detection of reactivity losses of the order of a few tenths of a percent in  $\Delta k/k$ .

Up to now no indication of possible dangerous situations arising from the use of this mode of reactivity control at power has been discovered during operation of power reactors.

However, as a conservative approach, a boron removal accident is usually studied for chemical shim PWR's, as the experience so far accumulated in the operation of these plants is not yet generally considered sufficient to give full assurance that a boron removal accident is out of the realm of credible events.

The present paper is intended to present and critically evaluate the available data on the phenomena connected with boron deposition and removal from core surfaces which may prove useful to the determination of the input reactivity for the study of a boron removal accident, namely, to the determination of maximum credible reactivity worth of removed boron in practical cases.

## 2 — PHENOMENA LEADING TO BORON DEPOSITION ON CORE SURFACES

Deposition of boron contained in the coolant of a pressurized water reactor with chemical shim on core surfaces may ideally take place by the following phenomena :

- a) boron deposition by phenomena of absorption by the crud deposited on core surfaces;

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(\*) Manuscript received on May 18, 1967.

- b) boron deposition due to the presence in the water of alkali metaborates. Alkali metaborates are usually present in the coolant of PWR's either because of the addition of potassium or lithium hydroxides or because of the build-up of lithium due to nuclear reaction of the boron. It can be imagined that precipitation of metaborates on core surfaces occurs owing to the following two phenomena : precipitation due to retrograde solubility of alkali metaborates (namely, decrease of solubility with increasing temperature) and then precipitation on the hottest parts of the core and, secondly, precipitation by concentration of metaborates on heat transfer surfaces due to distillation of water on them (it can be supposed that such a phenomenon takes place preferentially on crudded surfaces on which mass transfer between the surface of the fuel element and the coolant is deteriorated by the presence of the crud);
- c) boron precipitation on heat transfer surfaces by concentration of boric acid due to a phenomenon of distillation of the same kind as the one already mentioned in connection with alkali metaborates.

The evaluation of the amount of boron deposited on core surfaces implies, then, the evaluation of deposited crud, the evaluation of the amount of boron absorbed by deposited crud, the evaluation of alkali metaborate deposition and the evaluation of the amount of boron deposited according to the phenomenon mentioned under point c).

### 3 — EVALUATION OF THE AMOUNT OF CRUD DEPOSITED ON CORE SURFACES

To perform such an evaluation it is necessary to assume that all the crud released by the surfaces of the primary system is deposited on core surfaces, for no experimental results exist to evaluate the percentage of crud released by the primary system surfaces which is deposited on the core. It is, however, demonstrated that the crud deposits preferentially on heat transfer surfaces and in the course of some tests (WCAP 3729, Ref. 2) it has been noticed that practically all the crud released by metal surfaces is deposited on heat transfer surfaces. On this assumption the evaluation of the amount of crud deposited on the core is reduced to evaluation of the crud release rate by the primary system surfaces. This quantity times the ratio between total surface and core surface and times the operation time of the reactor will give the amount of crud deposited for unit core surface.

The available data on crud release rate are given in Table I.

The second column of the table contains the total corrosion rate; this quantity was measured after chemical descaling of the sample.

The third column contains crud release rates. This quantity is the one which is directly of interest for this evaluation; however, as can be seen, the pertinent data are less numerous than those which are available for the total corrosion rate (second column).

In the third column details are also given as to whether the numerical data have to be referred to the amount of released metal or of released crud. As the crud is essentially magnetite, the ratio between the amount of crud and the amount of contained metal is equal to about 1.4.

In the fourth column the duration of experiments is listed. This datum is very important for a weighted evaluation of the various experiments because of the strong dependence of the corrosion rate on the test duration; usually it can be observed that the corrosion rate decreases of some order of magnitude from the start of the test within a few hundreds of hours.

In the fifth column the number of the samples examined in the various experiments is listed.



The evaluation of the crud release rate of the Saxton reactor was performed on the basis of the measured amount of crud on the fuel elements and assuming a ratio between total surface and core surface of 2.5 and an operation time of four months.

The crud release rate of the Yankee reactor was evaluated on the basis of the amount of crud (tight plus loose) measured after 15 months operation (Ref. 7 — page 320, point 3), assuming a ratio between total surface and core surface of 3.5.

Among the data shown, those for experiments F and G are particularly high.

The data for experiment F are difficult to explain. As far as experiment G is concerned, the relatively short duration of the test may be the reason for the high result.

However, in view of the general trend of the data and particularly those, as for experiments H, I, L, which refer to tests performed with a water chemistry and test duration similar to those of interest for PWR's, a maximum figure of 3 mg/dm<sup>3</sup>/month seems a reasonable choice for the average crud release rate over operation periods of more than one year.

#### 4 — EVALUATION OF THE AMOUNT OF BORON ABSORBED BY CRUD

The evaluation of a theoretical maximum value of this quantity may be performed by a method proposed by Westinghouse (Ref. 1 — page 28 and following). An evaluation performed by this method is contained in Appendix A. The results show that for a natural crud the maximum boron content due to absorption is equal to 2280 ppm and for a typical synthetic crud 6850 ppm. The marked difference between natural and synthetic crud is due to the difference in exposed surface per unit weight for the two kinds of crud; typical values of this quantity are for natural crud 2.5 m<sup>2</sup>/g and for synthetic crud 7.5 m<sup>2</sup>/g.

The available data for absorption on crud are listed in Table II. It can be observed that the experimentally determined quantity of absorbed boron is in any case lower than the maximum theoretical amount calculated by the method described in Appendix A. This is true even in case c), in which, besides the boron absorbed on crud by exchange absorption phenomena, the amount of boron concentrated in crud by boiling of the solution has been considered.

#### 5 — BORON DEPOSITION DUE TO THE PRESENCE OF ALKALI METABORATES

As was mentioned in chapter 2, boron deposition due to the presence of alkali metaborates may possibly happen because of retrograde solubility and because of concentration on heat transfer surfaces due to boiling.

As far as the danger of metaborates precipitation due to retrograde solubility is concerned, it should be negligible because at the maximum temperature of the core heat transfer surfaces the solubility of these metaborates is still high (Ref. 11 — page 6-7; Ref. 3 — page 51).

The pertinent experimental data come substantially from two sources: the first is the out-of-pile loop described in Ref. 3 and the second the Saxton reactor.

The first experimental facility consisted in an out-of-pile loop with electrically heated rods. The rods were, alternatively clean or crudded, in the course of the tests. The cooling water contained a radioactive tracer (Na-22) whose concentration at heat transfer surfaces could be measured. Thermal fluxes and temperatures were of the same order of magnitude as those of a reactor. Several elements were tested in various thermal, hydraulic and chemical conditions.

The most important conclusions of these tests are the following :

1. A necessary condition for deposition is that nucleate boiling be present on heat transfer surfaces; as soon as the boiling was suppressed the concentration of solute on the surface was observed to disappear (reversible precipitation);
2. When the system NaOH-H<sub>3</sub>BO<sub>3</sub> was present in the water, no deposition was observed on clean heat transfer surfaces and the conclusion was reached that there is probably a lower limit of crud thickness below which no deposition occurs; such a limit would be of the order of magnitude of .3-.4 mill;
3. When, together with high crud thickness and the presence of nucleate boiling, a concentration of solute on heat transfer surfaces has been observed, the ratio between concentration on the surface and in the loop reached values as high as 8000 (Ref. 3 - page 60).

The experiments performed on this subject at Saxton may be divided into two parts.

The first part was performed in three stretches lasting up to ten days each. The aim of these tests was to determine the possible precipitation of potassium and lithium by measuring the concentration of these metals in the primary water. During the tests the purification circuit was isolated and the addition of pure water and chemicals discontinued.

The first test was performed in May-July 1963 with 3.5 ppm of potassium and 1200 ppm of boron in the water; the second test was performed in March 1964 with about 4 ppm of lithium and 500-600 ppm of boron; the third test was performed in March-April 1964 with 1.7 ppm of lithium and 450 ppm of boron. No deposition of alkali metals was detected within the limits of precision of the measurements. This accuracy, expressed as a variation in the boron concentration in the primary water, was  $\pm .026$  ppm boron for the tests with 4 ppm of potassium and  $\pm .0145$  ppm boron for the tests with 1.7 ppm of lithium. The tests performed at Saxton would have detected either reversible precipitation of alkali metaborates by concentration, found in the tests reported in WCAP-3731 (Ref. 3), or a possible precipitation due to retrograde solubility.

The second part of the Saxton experiments was aimed at the study of boron depositions due to nucleate boiling in the presence of large amounts of crud on core surfaces. It consisted in the injection into the primary coolant of several pounds of synthetic crud and subsequently in the study of the core reactivity behaviour in connection with the variation in the nucleate boiling in the core. These experiments were performed between November 1964 and January 1965. Alkali metaborates were present in the water. The test showed no reactivity effect connected with variations in the nucleate boiling.

## **6 — EVALUATION OF BORON DEPOSITIONS DUE TO BORIC ACID CONCENTRATION ON HEAT TRANSFER SURFACES**

The experiments just mentioned would have released depositions of boron due to this phenomenon if they occurred. As previously stated, no effect of this kind was discovered.

The Saxton test is probably the only one performed on this phenomenon. In the test reported in WCAP-3731 (Ref. 3) the possible concentration of boron due to boiling was not measured because the radioactive tracer used was sodium.

## **7 — EVALUATION OF THE PERCENTAGE OF THE DEPOSITED CRUD WHICH CAN BE RELEASED DURING THERMAL TRANSIENTS**

Available data on this subject come from three sources: the in-pile loop described in WCAP-3729 (Ref. 2), the Saxton reactor and the Yankee reactor. The above mentioned in-pile loop was particularly suitable for observation of massive crud releases because of the large amount

of crud deposited on fuel elements and because of the fairly high cooling rate allowed by the system (about 350 °F in 15 minutes, which corresponds to 23 °F per minute or to 13 °C per minute). Crud releases up to about 20 % were observed here.

In the Saxton reactor the maximum crud release was experienced during a loss of coolant flow test in October 1963; the amount of crud released was estimated to be of the order of 10 % of the total inventory deposited on core surfaces. Other minor crud releases were experienced during start-up and scram operations.

On Yankee reactor the magnitude of the observed crud released can be conservatively estimated to be 25 % of the deposited crud. These crud releases were experienced during start-up and scram with the first Yankee core.

The available data are given in Table III.

## 8 — EVALUATION OF THE MAXIMUM CREDIBLE REACTIVITY RELEASE IN A BORON REMOVAL ACCIDENT

On the basis of the previous considerations, the maximum reactivity addition in a boron removal accident may be evaluated as follows :

- a) Evaluation of the maximum amount of crud deposited on core surfaces. This amount may be obtained assuming a crud release rate of 3 mg/dm<sup>2</sup>/month from the surface of the primary system exposed to water and furthermore assuming that all the crud released by the primary surfaces is deposited on the core. With these two assumptions and knowing the operating life of the core, the amount of crud deposited on it can be calculated.
- b) Evaluation of the amount of boron absorbed by crud. This amount can be evaluated by the method outlined in Appendix A.
- c) From a) and b) the amount of boron deposited on the core is obtained. To evaluate the reactivity worth of it, the assumption of uniform distribution on core surfaces is not conservative. A factor should be applied to the reactivity in such a way as to account for possible disuniformities in distribution. As an example, a factor of two has been proposed for the Saxton reactor on the basis of observed crud distribution on fuel elements.
- d) Finally, to evaluate the maximum additional reactivity in case of accident, the percentage of deposited boron which is supposed to leave the core should be evaluated. Although the assumption that all the deposited boron is released seems overconservative, any other assumption would be arbitrary. Hundred per cent release is then suggested.

The reactivity ramp rate may then be evaluated assuming that the released boron leaves the core at the velocity of the cooling water.

In practical cases an evaluation performed as outlined above leads to released reactivity amounts of a few tenths of a percent  $\Delta k/k$  in about half a second.

On the basis of the available information, boron deposition due to retrograde solubility of alkali metaborates and to concentration phenomena connected with nucleate boiling seems very improbable and has been disregarded in the previous evaluation.

This assumption, however, should be substantiated by the result of tests to be performed during operation and intended to detect the possible effect of nucleate boiling on core reactivity. Because the nucleate boiling boron concentration phenomenon would be dependent on the amount of deposited crud and because this amount increases with time, such a test should be periodically repeated during the core life.

TABLE I

## Data on high-temperature corrosion rates of AISI 304 and 316 stainless steels

Source	Total corrosion rate (mg/dm <sup>2</sup> /month)	Release rate (mg/dm <sup>2</sup> /month)	Test Duration (hr)	Number of samples	Notes
A. Ref. 4 p. 101 & foll.	5 ÷ 15	—	1500-5000	Several tens	(1)
B. Ref. 5 p. 307 & 311	5 ÷ 10	2 — 3 (probably metal)	—	—	(2)
C. Ref. 5 page 311 Ref. 8	—	.2 (probably metal)	—	—	(3)
D. Ref. 6 - p. 21	5 ÷ 10	2.3	—	—	
E. Ref. 1	3.5	0	2360 — 2680	—	(4)
F. Ref. 2	20 ÷ 55	10 — 35	2400	8	(5)
G. Ref. 12	21.8	12.5 (crud)	1440	10	(6)
H. Ref. 13	5.65	3.22 (crud)	6600	7	(7)
I. Saxton	—	< 1.6 (crud)	9 months in the reactor, 3000 hr at full power	4 fuel rods	
L. Yankee Ref. 7 Page 320.3	—	< 1.7 (crud)	15 months in the reactor	1 fuel rod	(8)

## Notes to Table I :

(1) Out-of-pile.

(2) Out-of-pile.

(3) Out-of-pile; pH = 9.8 — 10.2 at 25 °C by addition of LiOH and KOH.

(4) Out-of-pile test in static conditions (no flow).

(5) In-pile (WTR).

(6) Out-of-pile; 304 SS specimens; partially neutralized boric acid solution (1050 ppm B; 1.6 ppm Li) at 600 °F; flow velocity 6 — 20 fps.

(7) Out-of-pile; 304 and 316 SS specimens; partially neutralized boric acid solution (1550 ppm B; 5.7 ppm K) at 600 °F; flow velocity 6 — 20 fps.

(8) The fuel rod to which reference is made here is the only one which was not brush cleaned before chemical attack and therefore the quoted value of the crud release rate (1.7 mg/dm<sup>2</sup> month) includes both the loose and the tight crud; on other fuel rods only the tight crud was measured, after chemical descaling, and the result was about 1.5 times lower. Care was exercised during transportation of the fuel elements from the reactor to the hot cells to avoid any undesired crud detachment before examination.

TABLE II

Data on exchange absorption of boron on crud

Source	Kind of crud	Radiation		Test Duration	Boron in solution ppm	Temperature (°C)	Number of specimens	Boron in crud (ppm)	Notes
		with	without						
A. Ref. 1	Synthetic		x	7 days	1100	25	8	900	(1)
	Synthetic		x	24 hr	1400	310	7	3600	(2)
B. Ref. 2	Natural	x		3.4 months	1500	310-345	4 fuel rods	200—300	(3)
	Natural	x		3.4 months	1500	310	3	500—600	(4)
C. Ref. 10 page 13	Synthetic	x		20 days	~ 900	200	1	6500	(5)
D. Yankee Ref. 10 page 19	Natural	x			400	~ 280	1	1200	(6)
E. Saxton	Natural	x		9 months	~ 1200	~ 280	4 fuel rods	200—900	(7)

Notes to Table II :

- (1) Oscillating pyrex containers; typical result.
- (2) Stationary autoclave; in this experiment as in the previous one the mixture crud-solution was centrifuged after the test and the amount of absorbed boron determined by measurements of solution concentration and concentration in the centrifuged solid. In this case also a typical result has been inserted in the table.
- (3) The rods were washed for a long time after exposure in order to remove decay heat.
- (4) Circulating, not washed crud.
- (5) The high value of the absorbed crud may be explained by the test conditions; in particular the figure of 6500 ppm is inclusive of the boron concentrated in crud by distillation due to boiling.
- (6) Circulating, not washed crud.
- (7) The rods were probably washed before sampling.

TABLE III

Percentage of crud release

Source	Released crud %	Notes
A. Ref. 2 In-pile loop	14.6	Start-up
	2.4	Cool down (23 °F/min)
	19.4	Cool down (23 °F/min)
B. Saxton	10	Loss of flow test
C. Yankee	25	Start-up
	25	Scram

## APPENDIX A

### Evaluation of a theoretical upper limit for the boron absorbed by crud

*Principal assumptions :*

1. Crud is essentially magnetite,  $\text{Fe}_3\text{O}_4$  (Ref. 4, page 230 and Table 12-3) and the active surface per unit weight is  $2.5 \text{ m}^2/\text{g}$  for a natural crud and  $7.5 \text{ m}^2/\text{g}$  for a synthetic crud (assuming for natural crud a particle diameter of .5 micron (Ref. 4, page 231) and for magnetite a density equal to 5.18, the calculated active surface per unit weight  $2.3 \text{ m}^2/\text{g}$ ).
2. The mechanism of absorption consists in the passage of the ferrous ion of  $\text{Fe}_3\text{O}_4$  to ferric and in the consequent exchange of the resulting free valence with that of a trimer ion of boric acid (one valence associated with three boron atoms).

With the assumption that magnetite has a cubic structure, the area of one face of the molecule,  $A_m$ , is :

$$A_m = \left( \frac{231.52}{6.023 \times 10^{23} \times 5.18} \right)^{2/3} = 1.77 \times 10^{-15} \text{ cm}^2$$

The surface per unit weight,  $S_c$ , is, by assumption,  $2500 \text{ cm}^2/\text{g}$  for natural crud and the number of active molecules per unit weight of crud,  $N$ , is :

$$N = \frac{S_c}{A_m} = \frac{2.5 \times 10^4}{1.77 \times 10^{-15}} = 1.41 \times 10^{19} \text{ active molecules/g}$$

3. Weight of boron absorbed by an active molecule.

As it is assumed that each Fe atom of an active molecule of crud absorbs three atoms of natural boron (atomic weight = 10.82), the weight of boron per active crud molecule,  $P_{BM}$ , is :

$$P_{BM} = \frac{3 \times 10.82 \times 3}{6.023 \times 10^{23}} = 1.62 \times 10^{-22} \text{ g/molecule}$$

The maximum theoretical weight of boron absorbed by the unit weight of crud,  $P_{BT}$ , is then, for natural crud :

$$P_{BT} = 1.62 \times 10^{-22} \times 1.41 \times 10^{19} = 2.28 \times 10^{-3} \text{ g/g} = 2280 \text{ ppm.}$$

For a typical synthetic crud ( $S_c = 7500 \text{ cm}^2/\text{g}$ ) :

$$P_{BT} = 6850 \text{ ppm.}$$

## APPENDIX B

### Useful data for evaluation of maximum reactivity release

It is assumed that the ratio between the total primary surface area and the core surface area is 3.5 and that 100 ppm of boron in the primary water are equivalent to 1 %  $\Delta k/k$ .

- 1 mg crud/dm<sup>2</sup> corresponds to a deposit of 0.0025 mill thickness (density of magnetite = 5.18; crud porosity = 0.7, which is the maximum value found in the literature, Ref. 3, page 60).
- 1 mg boron/dm<sup>2</sup> of core surface corresponds to .4 %  $\Delta k/k$ .
- If 1 ppm boron is removed from the primary water and deposits on core surfaces, such a deposition is equivalent to an increase in the boron concentration in the core of 16 ppm.
- A crud release of 1 mg crud/dm<sup>2</sup> month from the primary surfaces corresponds to a crud increase in the core of  $\sim 3.5$  mg/dm<sup>2</sup> month (it being assumed that all the crud is deposited on core surfaces).

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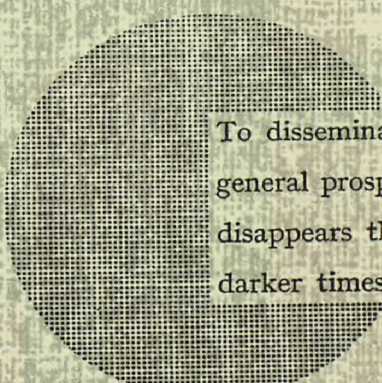
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Alfred Nobel

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