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Information from the Water Ingress
Accident on AVR

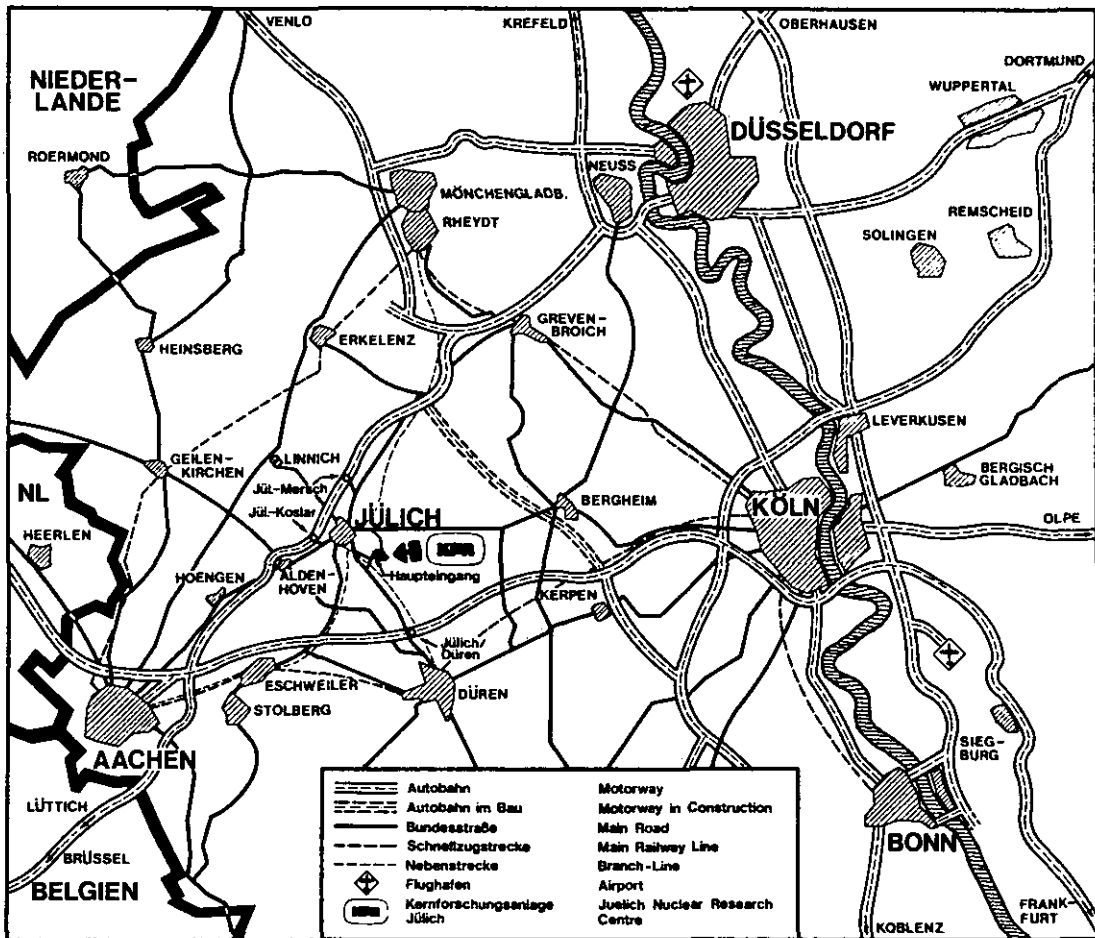by

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Zusammenfassung

Im Mai 1978 ereignete sich ein Wassereinbruch im AVR. Der Reaktor war abgeschaltet und wurde durch Zwangsumlauf gekühlt. Wasser drang aus einem Überhitzerleck in den Primärkreislauf ein, verdampfte beim Durchgang durch das Core und kondensierte im unteren Teil des Primärkreislaufs und im Bereich des Kugelabzugs.

Im Wasser wurden verschiedene Spaltprodukt-Aktivitäten von den Wissenschaftlern des AVR gemessen. Eine Studie wurde begonnen, um die Quellen der Aktivitäten zu identifizieren und Informationen zu gewinnen, die allgemein bei der Analyse von Wassereinbruch-Störfällen benutzt werden können.

Der erste Teil dieser Studie ist in diesem Bericht dargestellt. Die möglichen Aktivitätsquellen werden einzeln betrachtet und ihre Beiträge abgeschätzt, wobei Daten aus früheren Laborexperimenten - soweit möglich - herangezogen wurden.

Das Hauptergebnis ist, daß wertvolle Informationen hinsichtlich der Desorption von Jod, Cäsium und Strontium von dem Staub und den Oberflächen des Primärkreislaufs gewonnen worden sind. Es wurde ein Minimal-Programm von Messungen und analytischen Arbeiten erstellt, die notwendig sind, um diese Informationen zu ergänzen. Ein Beispiel der Anwendung dieser Daten auf einen speziellen Leistungsreaktor-Störfall wird gegeben, um zu zeigen, wie diese Informationen die Berechnung der Auswirkungen beeinflussen kann.

Für den zweiten Teil der Studie sind bessere Abschätzungen der Spaltprodukt-Konzentrationen im Primärkreislauf vor dem Unfall sowie verschiedene Messungen erforderlich, die auszuführen sind, wenn der Reaktor wieder in Betrieb ist.

Information from the Water Ingress Accident on AVR

Contents

Summary

An ingress of water occurred in the AVR reactor in May 1978. The reactor had been shut down and cooled by forced circulation; liquid water entered the primary circuit from a leak in the superheater, evaporated in passing through the core and condensed in the lower part of the primary circuit and in the ball handling region.

Various fission product activities were measured in the water by the AVR scientists and a study was started to identify the sources of these activities and to derive information which could be used in the analyses of water ingress accidents in general.

The first part of this study is reported in this note. The possible source terms are considered separately and estimates of their contributions are made, supported by data from previous laboratory experiments where possible.

The main conclusion is that valuable information has been derived concerning the desorption of iodine, caesium and strontium from dust and primary circuit surfaces. A minimum programme of measurements and analytical work necessary to increase this information has been identified. An example of the application of the data to a particular accident to a power reactor is given to indicate how the information can affect the calculation of consequences.

For the second part of the study, better estimates of the fission product concentrations in the primary circuit prior to the accident and various measurements when the reactor is operating again are required.

Information from the Water Ingress Accident on AVR

1. Objectives

In principle, data derived from the water ingress accident can provide unique information in the following areas:

- the transport of heavy metal oxides or carbides by corrosion or leaching mechanisms;

- the effect of abnormal water concentrations on fission products external to the coated particles i.e. fission products deposited in the graphite of the fuel elements, on dust or on circuit surfaces;

- the effect of water on defective fuel particles.

Currently the available experimental data in these areas are sparse and large differences exist in the assumptions used in analysing the effects of water ingress. For example, in the Accident Initiation and Progression Analysis carried out by the General Atomic Company (1), no account was taken of the possibility that steam could cause desorption or wash-off of fission products deposited on primary circuit surfaces. On the other hand, a prediction made at KFA (2) of the release of activities from the AVR following steam ingress includes an assumption that 50 % of the metallic fission products and 80 % of the iodines deposited in the primary circuit would desorb immediately.

This difference in treatment can change the contribution to the risk from water ingress accidents from negligible to significant levels particularly because the deposits of long-lived metallic fission products increase throughout the life of the reactor.

The information from the AVR accident cannot be precise because the conditions were not under control but at least gross assumptions can be examined, order of magnitude estimates can be deduced and in some areas certain effects can be eliminated.

2.   Sources of Fission Products

The course of the accident was an injection of water in the liquid phase from a superheater tube over several days which evaporated in passing through the sub-critical core and condensed in the ball handling region and the lower part of the primary circuit. Various fission product activities were measured in this water (3); the results are reproduced in Table 1.

The possible sources of these activities are:

2.1 - removal of fission products from defective fuel particles;

2.2 - corrosion of core graphite containing fission products;

2.3 - leaching of fission products from fuel balls submerged in water;

2.4 - leaching or desorption of fission products from the graphite of fuel balls in the core;

2.5 - leaching or desorption of fission products from dust or graphite fragments;

2.6 - wash-off or desorption of fission products deposited on primary circuit surfaces;

2.7 - exchange of tritium adsorbed on graphite.

3.   Estimates of the Source Terms

3.1   Removal of Soluble Fission Products from Defective Particles

There was no trace of the gamma activities which would be directly associated with damaged fuel particles.

These activities include: proto-actinium 233, cerium 141/144, zirconium/niobium 95, ruthenium 103/106.

These species have low diffusion coefficients relative to caesium or barium and do not migrate in normal operation from the immediate neighbourhood of the fuel kernels. A direct experiment to test the

transport of fission products was made by exposing irradiated fuel compacts to high steam concentrations at 1250° C until the compacts disintegrated (4). Several of these species were detected together with caesium, iodine and xenon even though the defective particle fraction was as low as 10^{-5}.

The absence of these activities in the AVR water indicates that damaged fuel particles can be eliminated as a dominant source term for the metallic isotopes - which is an important conclusion when the extremely small fraction of released fission products is considered in comparison with the total inventory in the core.

3.2 Corrosion of Fuel Element Graphite

Any corrosion which occurred during the passage of steam through the core was in the chemical regime because the graphite temperatures did not exceed 500° C. The steam concentration was uniform throughout the fuel element material and any corrosion was uniformly distributed.

The weight loss has been estimated by Moormann (5) assuming a water concentration of 6000 vpm for 15 days with all the core graphite at 500° C and the reactor pressure at 10 bar.

The predicted weight loss was 0.8×10^{-4} mg cm^{-2}, i.e. the fictional equivalent weight loss if all the corroded graphite had been removed from the surface. This is important because the caesium and strontium concentrations have been found to be higher at the surfaces of fuel balls with high burn-up than in the interior (6, 7). Therefore any estimates based on surface corrosion will be upper limits. The measurements made on a fuel element which had received a burn-up of 17.6 % fima show surface concentrations of strontium 90 of 1×10^{17} atoms/gram graphite and of caesium 137 of 2×10^{14} atoms/gram graphite.

If these are assumed pessimistically to be the surface concentrations throughout the core then the expected maximum removal of strontium 90 activity is given:

$$A_{sr} = 10^{17} \times 4\pi r^2 N \times m \times \frac{\lambda}{3.7 \times 10^{10}} \quad \text{curies}$$

N = number of fuel elements in the core (10^5)

r = ball radius   cm

m = mass of graphite removed per unit surface area gcm^{-2}

λ = decay constant sec^{-1}

giving A_{sr} = 1.8 milli Curies

and for caesium 137,

A_{cs} = 3.4 micro Curies.

The activities released by corrosion of the fuel elements are therefore negligible.

3.3 Leaching of Fission Products from Submerged Fuel

The specific activities of caesium 134, caesium 137 and strontium 90 in the water contained in the 4 control rod guide tubes and in the water covering the discharged fuel were the same within experimental error. These two quantities of water were separated by the guide-tubes up to the top of the fuel discharge tube. Therefore the submerged fuel elements in the fuel discharge system did not contribute to the release of metallic fission products. This also means that the dissolving by liquid water of active species adsorbed or diffused into graphite is a long term process. There is qualitative confirmation from reported measurements of fission products leached from irradiated fuel balls stored in a sealed can into which water leaked over a period of about one year (8). The measured removal of caesium 137 was 380 μ Ci from 49 balls stored in the can, corresponding to a removal fraction of approximately 4×10^{-6}. Applying this fraction to the 5000 balls in the discharge system, the expected removal of caesium 137 would be about 60 mCi, with no allowance for time dependence of solubility.

The caesium and strontium activities from this source are therefore negligible.

3.4 Removal of Fission Products from the Fuel Element Graphite by Desorption in Steam

Because the water passed through the core as saturated steam at 500° C, any significant release of fission products from the fuel elements would be a desorption process affected by the high water concentration. In dry helium the desorption rates of caesium and strontium from graphite at 500° C are negligible. For caesium, for example, the level of 2×10^{14} atoms/gram graphite corresponds to a concentration of 0.05μ gm caesium/gram graphite. At this concentration, the adsorption isotherm is Langmuir and the evaporation rate would correspond to a partial pressure of $\sim 10^{-18}$ bar i.e. approximately 10 atoms caesium /cm^3 helium (relatively trivial).

There is no reported evidence of the effect of steam on caesium diffused into graphite. Experiments, however, were made to determine the catalytic affects of caesium, barium and strontium on the corrosion rates of graphite in steam at 25 mbar and at temperatures between 700° and 1000° C (9). The metal loadings were higher than those of the AVR graphite by orders of magnitude but no displacement of those metals was observed. This is only a qualitative indication that fission product metals which have diffused into graphite may not be affected by steam. Further evidence is necessary to determine the relative importance of this source term, particularly with respect to strontium.

3.5 Leaching and Desorption from Dust and Primary Circuit Surfaces

3.5.1 Iodine

Within the information available, desorption from dust and from circuit surfaces cannot reliably be distinguished and these two sources are discussed together.

The equilibrium concentration of iodine 131 in the AVR core (46 MW$_t$) is 1.3×10^6 curies. Assuming an effective R/B of 10^{-5} and a plate-out factor of 10^4, which are typical for an HTR, the equilibrium concentrations in the coolant and on the circuit surfaces would be 1.3 mCi and 13 Ci respectively. No iodine remains in the core graphite.

The adsorption isotherm for iodine on the AVR boiler steel shows that less than 1 % of the plated-out iodine would be deposited on the super-heater section, and wash-off by water had, therefore, little effect.

The gas-borne activity was negligible and the majority of the iodine measured in the water must have been desorbed from the primary circuit surfaces and dust downstream of the superheater.

The fraction desorbed in a helium/steam mixture can be estimated from the results of experiments made at ORNL (10). Müller (11) has derived the desorption equation:

$$A_{desorb} (t) = A_{p.o.} (o) \frac{K}{\lambda + K} \cdot \left[1 - e^{-(\lambda + K).t} \right]$$

where A_{desorb} (t) is the time dependent desorption of iodine activity in Ci.

$A_{p.o.}$ (o)   is the plated-out activity of iodine at time zero in Ci.

λ decay constant of iodine sec^{-1}

K iodine desorption coefficient sec^{-1}

Inserting the values of K derived from the ORNL experients and extra-polating to the temperature conditions during the AVR accident, the desorbed fraction at 100 hours is 80 %, which is in good agreement with the desorption fraction assumed by von der Decken et al. (2).

However, the estimated level of iodine 131 in the AVR primary circuit immediately prior to the water ingress is 10 Ci (12) and the measured activity in the water was 1.4 Ci, corresponding to a desorption factor of only 14 %.

It is therefore important to check the core release rate and the circuit inventory of iodine prior to the accident.

3.5.2 Caesium

An approximate estimate of the caesium 137 activity in the circuit immediately prior to the ingress is 20 Ci, based on the estimated level

of von der Decken (2) and the integrated release from fuel failures sub-
sequent to that level given by Maly et al. (13). The measured release
of 1.8 Ci would then correspond to a desorption fraction of 9 %.

Wahl and Jacobsen (3) have pointed out that the Cs 137 / Cs 134 activity
ratio in the water (1.84) corresponds more closely to the ratio found
in the dust filtered from the gas (2.08) than to the ratio,measured on
fuel element surfaces (1.24). No reliable estimate of the desorption
factor can be made until a calculation of the levels and distributions
of the caesium isotopes in the AVR circuit prior to the accident has been
carried out, based on Vampyr loop measurements.

3.5.3 Strontium

The high level of strontium 90 activity in the water represents the
most important measurement of the incident. The Sr 90 / Sr 89 ratio of
100 indicates an abnormally high release of strontium occurring 3 - 4
years ago. This is supported qualitatively by the strontium concentration
profiles on high burn-up fuel balls where the internal diffusion profiles
represent the history of the individual ball and the profiles from the
surface inward represent the history of the reactor.

As for caesium, a desorption factor cannot be estimated until a calculation
of the strontium concentrations in the primary circuit has been made.

3.6 Tritium

The measured concentrations of tritium in normal operation are
3×10^{-9} Ci/N cm^3 in the helium and 3×10^{-4} Ci/cm^3 in the graphite of
both fuel elements and graphite balls (15), leading to estimated tritium
inventories in the graphite of the core and of the submerged balls of
3400 Ci and 170 Ci respectively. Approximately 20 % of this tritium was
accessible for an immediate exchange with hydrogen in the water; above this
fraction, a temperature dependent diffusion process is rate limiting and
at the temperatures of the fuel elements during the accident, no further
significant contribution from the fuel graphite and none from the coated
particles is to be expected.

This source would therefore account for about 700 - 800 Ci of the 2900 Ci measured in the water and if an exchange model with a maximum of 20 % accessible tritium is used to account for the remaining 2200 Ci, a reservoir of the order of at least 10 000 Ci must be assumed on the reflector and structural graphite.

The distribution model proposed by ORNL for evaluation of the Fort St. Vrain measurements (16) includes the following assumptions:

- that 50 - 70 % of tritium produced in the coolant from He 3 is chemi-sorbed on graphite,

- that all the tritium produced by Li 6 (n, α) reactions is retained within the graphite.

The He 3 source in AVR produces not more than 100 Ci/a and probably closer to 50 Ci/a which would account for 300 - 500 Ci of chemi-sorbed tritium.

The production in the graphite must be a guess because the lithium and boron concentrations are not known. However, the measurements made on a Dragon reflector block showed a tritium concentration of 120 μ Ci/g at the inner surface, diminishing to 50 μ Ci/g at 5 cm depth (17). A very approximate extrapolation of this profile to the AVR reflector allowing for the different neutron dose leads to an estimated total of 1500 Ci for a Li 6 impurity concentration similar to that of the Dragon block i.e. 0.03 p.p.m. If the lithium impurity is as high as 0.3 p.p.m., a tritium reservoir of 10 000 - 15 000 Ci in AVR is a possibility from this source.

The necessary work to justify these estimates are:
- measurements of the lithium concentration in AVR reflector graphite,
- measurement of the tritium levels in the fuel free zone of elements now in the core,
- injections of hydrogen during operation to measure the tritium now accessible for exchange,
- more rigorous calculations of the tritium inventories prior to the accident.

This data will be valuable for the calculation of tritium balances in the THTR and in the advanced reactors.

4. Application of the Data to Accident Analyses

The application of the data from AVR is best shown by taking an example.

In the Phase II Report of the AIPA Study (14), a water ingress accident is analysed in which the risk of introducing a design option to reduce cost is estimated. The design option involves the elimination of dump tanks so that the steam generator inventory is released directly to the atmosphere, by-passing the containment. The risk relates to the failure to close this by-pass line after dumping the steam, thus causing a depressurization of the reactor direct to atmosphere. The concern here is not with the probability of the event but with the consequences.

The fission product source terms are shown in Table 2 for three cases:

- the values used in the AIPA Study

- the values modified by assuming a 15 % desorption factor for strontium, caesium and iodine (which might be the outcome from AVR)

- the values modified by assuming a 50 % desorption factor for strontium and caesium and 80 % for iodine (which are the assumptions used by von der Decken et al. (2)).

Taking account only of the increased contribution to the release of iodine 131, the effect on the 30 day thyroid dose is an increase from 1 rem to 15 rem (taking 15 % desorption) and to about 70 rem (taking 80 % desorption). The latter figure may be pessimistic but is much closer to the reality of the AVR accident than the assumption of AIPA.

The list of source terms also underlines a further effect which would be apparent when the AVR reactor is re-started. This is the possible reaction of steam with defective carbide fuel. This reaction transforms uranium and thorium carbides to the oxides, and if enough steam is present, hydrate molecules can be attached to the oxides. This hydrolysis causes a swelling of the particles and the release of gaseous fission products. Hydrolysis is zero above 700° C and increases with decreasing temperature (14). In the conditions of the AVR accident,

any defective carbide fuel is likely to have been hydrolysed and it is
necessary to operate the reactor at full power and at the prior
temperatures in order to measure any overall increase in fission gas
release.

5.   Proposals for Further Work

The two areas of information which can be of direct value to accident
analyses of HTR systems are:

- the desorption factors for iodine and metallic fission products,

- the extent of hydrolysis of defective carbide fuel.

The tasks required to obtain these data are:

- the estimation from Vampyr loop data of the levels and distribution
  of activities deposited on primary circuit surfaces and on dust
  immediately prior to the accident including in particular:

caesium 134	strontium 89
caesium 137	strontium 90
silver 110m	iodine 131

- the measurement of individual rare gas isotopes in the coolant as a
  function of time when the reactor is operating at the same conditions
  as those immediately prior to the accident for which gas activity
  measurements are available for isotope-to-isotope comparison.

Other information which would be of value in understanding transport
mechanisms include:

- check of the Cs 134 / Cs 137 and Sr 89 / Sr 90 ratios in activities
  deposited on any primary circuit component which may be removed for
  inspection. Also, the same ratios for dust samples if these are taken
  from the circuit;

- a direct experiment on high burn-up balls removed from the reactor
  several months after re-start to determine the desorption factors of
  strontium 90 and 89 from fuel ball surfaces in a saturated steam
  atmosphere at temperatures from 500° C to 700° C. This is probably
  specific to AVR fuel because the strontium loadings reported
  (15 μ gram Sr / gram graphite) is abnormally high;

- injection of hydrogen when the reactor is at power to check tritium release per mol of injected hydrogen;

- post-irradiation examination of fuel with carbide kernels for hydrolysis damage;

- post-irradiation examination of tritium levels in fuel ball graphite from balls now in the reactor core;

- post-irradiation examination for any significant changes in surface concentrations and profiles in the fuel-free zone of fuel balls now in the core compared to previous measurements;

- check of any change of the fission gas release levels immediately before and after the injection of 8 kg of water in March 1978.

6.   Conclusions

The water ingress accident on AVR has provided unique information with regard to the sources of fission products which must be considered in the analysis of such accidents for HTR's.

It has been shown that upper limit estimates can be made for these sources and that several can be eliminated. In other cases, further work is required to reduce the uncertainties of the size of the contributions. In particular the removal of strontium from dust, from primary circuit surfaces or from fuel element graphite cannot be defined with confidence on present information.

7.   Acknowledgements

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| | drainage of the first 2000 l water | | | | | water tank 21000 l 06.07.78 | 4100 l water in tubes of shut-down rods 13.09.78 | |
	01.06.78	07.06.78	08.06.78	09.06.78	10.06.78			13.05.78
Cs 134	0.173	0.0737	0.0813	0.0849	0.0923	0.0387	0.0388	0.972
Cs 137	0.24	0.123	0.128	0.135	0.152	0.0705	0.0736	1.782
Sr 89	n.m.	0.007	0.022	0.16	0.089	0.014	n.m.	0.351
Sr 90	n.m.	7.15	1.99	2.03	1.62	1.46	~1.3	36.65
J 131	0.12	0.055	0.054	0.061	0.058	0.056	d.n.d.	1.41
H 3	170	n.m.	110	110	100	115	120	2907.
Eu 154	0.07	0.017	0.0013	0.0017	n.d.	n.d.	n.d.	-
Eu 155	0.05	0.01	0.001	0.0013	n.d.	n.d.	n.d.	-
Eu 156	0.76	0.16	0.011	0.022	n.d.	n.d.	d.n.d.	-
Co 60	0.0007	n.d.	n.d.	n.d.	n.d.	0.00004	0.00015	0.0015
Zn 55	0.001	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	-

Table 1: Activity concentrations in the leakage water

n.m. ≙ not measured   n.d. ≙ not detectable   d.n.d. ≙ decayed, not detectable

Table 2.   Fission Product Source Terms for a Water-Ingress Accident

on the GAC - 1160 MW HTGR Design

	Xe + Kr Isotopes (Ci)	Cs 137 (Ci)	Sr 90 (Ci)	I 131 (Ci)
AIPA Study				
1.				
Activity released by corrosion	-	225	28	-
Activity released by hydrolysis of fuel	8 x 10⁵	-	-	-
Activities on dust lifted by depressurization	-	0.3	-	89
Totals	8 x 10⁵	225	28	89
2.				
15% desorption of caesium, strontium and iodine plated out on surfaces or dust	-	6,100	52	1,230
Totals	8 x 10⁵	6,325	80	1,319
3.				
50% desorption of caesium and strontium and 80% desorption of iodine plated out on surfaces or dust	-	20,000	173	6.670
Totals	8 x 10⁵	20,225	201	6,759