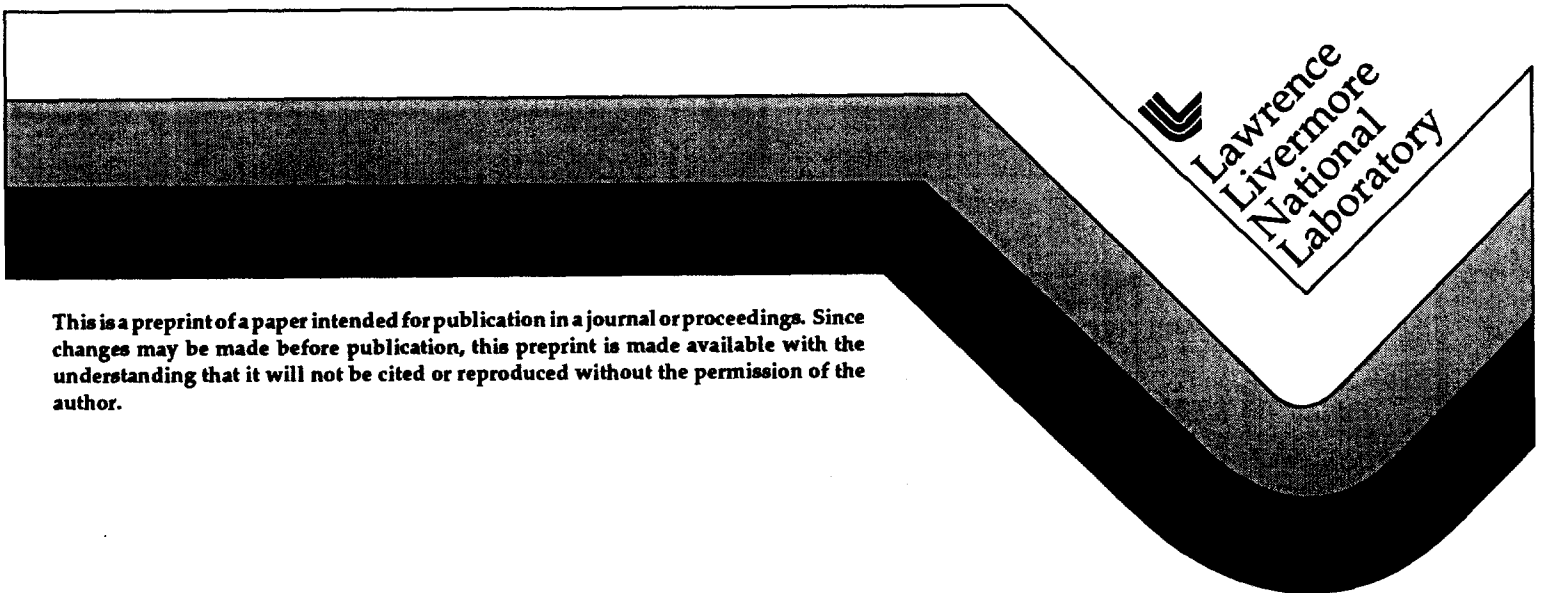


**Possible Criticality of Marine Reactors Dumped
in the Kara Sea**

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POSSIBLE CRITICALITY OF MARINE REACTORS DUMPED IN THE KARA SEA

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Abstract

The largest inventory of radioactive materials dumped in the Kara Sea by the former Soviet Union comes from the spent nuclear fuel (SNF) of seven marine reactors. Using corrosion models derived for the International Arctic Seas Assessment Project (IASAP), the possibility of some of the SNF achieving criticality through structural and material changes has been investigated. Although remote, the possibility cannot at this stage be ruled out.

Introduction

In the Spring of 1993, an English translation of the Russian report, "Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation" (Yablokov et al, 1993) was released. The findings presented in this report were the result of a scientific study commissioned in October 1992 by the Office of the President of the Russian Federation. The White Book, as the report was later called, reported that sixteen marine reactors from seven former Soviet Union submarines and the nuclear icebreaker *Lenin* had been dumped at five sites in the Kara Sea. Six of the submarine reactors still contained spent nuclear fuel (SNF), the term used to describe uranium fuel in which some of the isotope responsible for most of the energy production, ²³⁵U, had been fissioned (split apart) into daughter radionuclides. SNF from the centreline reactor of the icebreaker *Lenin* had also been placed in a container and dumped separately from its reactor pressure vessel (RPV). The locations of the dump sites, all on or near the coast of the Arctic island of Novaya Zemlya, are shown in Figure 1.

International concern over the possible health and environmental effects from disposal of these reactors and other radioactive wastes in the shallow waters of the Arctic Seas prompted the International Atomic Energy Agency (IAEA), as part of their responsibilities to the London Convention of 1972, to initiate the International Arctic Seas Assessment Project (IASAP) (Sjoebloom & Linsley 1993). As part of that project, the Source Term Working Group was tasked to predict radionuclide release rates for all the dumped reactors, generically known as steam generating installations (SGIs), which required analysis of the corrosion processes and timescales within the SGI components. As well as allowing prediction of radionuclide release rates, these analyses also allowed some rough estimates to be made of whether the reactor core could achieve criticality as corrosion progressed. A reactor is said to be critical when the neutron chain reaction, fuelled by ²³⁵U, is self-sustaining. Achievement of criticality is the goal of all reactor designs, as it enables the reactor core to produce a constant amount of energy by the fission of successive ²³⁵U nuclei. In the case of the dumped SGIs, there was concern that further criticality could lead to accelerated corrosion and radionuclide release rates, production of new radionuclides, and, in the worst case, explosion and structural disintegration of the SGI.

The results and conclusions of the Group are to be published in an IAEA Technical Report "Predicted Radionuclide Release from Marine Reactors Dumped in the Kara Sea,; report of the Source Term Working Group of the International Arctic Seas Assessment Project (IASAP)" (IAEA 1997). This paper expands on the discussion of criticality in that report; it first describes the condition of the cores as it affects criticality, and then makes some preliminary calculations to assess the possibility of criticality occurring. No attempt is made to quantify such possibility except in the broadest terms.

Reactor Operating Histories & Disposal

In all cases, the dumped SGI was taken out of service owing to possible or actual core damage after an accident. Details of operating histories, including fuel burnup in GWd, were made available to the IASAP, enabling calculation of the amount of ^{235}U left in each core.

The White Book referred to each SGI solely by the hull factory number used during build. Nilsen et al (1996) correlated factory number with submarine type, allowing identification of the submarine and the accident resulting in reactor disposal. Table 1 gives a summary of the dumped units and accident descriptions, and Table 2 gives details of fuel load, reactor type and operating history for each SGI.

Disposal operations of the pressurised water reactors (PWRs) were described by Sivintsev (1993, 1994). With one exception, all nuclear submarine PWRs containing SNF were dumped in their separated reactor compartments (RCs). The SGI from submarine factory number 421 was removed from its RC and placed in a steel and concrete structure on a barge, before the whole assembly was dumped in the Novaya Zemlya Depression. Before disposal, the primary circuit loops and equipment were washed, dried, and sealed. The RPVs, which still contained SNF, were filled with a patented hardening compound, labelled Furfurol(F), based on furfural, an organic resin. The control rods were left fully inserted in the cores, welded in place and the control rod channels filled with Furfurol(F) and sealed with 10 mm stainless steel (SS) caps.

The three PWRs of the icebreaker were discarded within their RC (Sivintsev 1995b). The centreline (N2) reactor was too badly damaged to be completely defuelled, so after partial defuelling the whole core barrel of the N2 reactor was removed with 60% of the SNF in place. This core barrel was sealed in a SS container known as Container C, filled with Furfurol(F), and dumped close to the icebreaker RC.

The submarine containing the two liquid metal reactors (LMRs) suffered a secondary to primary leak in the left board reactor whilst on patrol; this caused severe core damage and led to approximately 20% of the fuel from the left board core being transported to the steam generators (Giltsov et al 1992; Yefimov 1994b). The right board core remained intact. Prior to dumping, a number of actions were taken to secure the LMRs for disposal including the use of some 2 m³ of Furfurol(F) and 250 m³ of bitumen (Yefimov 1994a). The rod channels were filled with Furfurol(F) prior to dumping, and sealed with 10 mm SS caps. Both LMRs were then dumped in the complete submarine.

At the time of disposal, all the RCs were allowed to flood, thereby exposing any unprotected external surface of each RPV to sea water.

Core construction

It is important to describe the core construction, as this has a large effect on the possibility of criticality. The construction and layout of the submarine and icebreaker PWRs are described by Sivintsev (1994), and the LMRs contained in the sunken experimental November class submarine factory number 601 by Yefimov (1994a,b).

Each PWR consisted of a cylindrical carbon steel RPV with approximate dimensions: 1.4- to 2-m diameter, 3.4- to 5-m height, 100- to 120-mm thick walls. It was assumed by the Group that nuclear submarine cores were loaded with U-Al alloy fuel containing 50 kg of ^{235}U at an enrichment of 7.5% or 20%; natural uranium contains around 0.7% ^{235}U . Cores in the icebreaker were loaded with varying quantities of UO_2 sintered ceramic fuel enriched to 5.0% ^{235}U and clad in Zr-Nb alloy or SS. Information about the number and structure of the control rods was unavailable for any of the submarine PWRs; 10-mm radius boronated SS rods were assumed.

Each LMR consisted of a cylindrical SS RPV with approximate dimensions: 1.8-m diameter, 3.7-m height, and 30-mm thick walls. LMR cores were loaded with 90 kg of ^{235}U enriched to 90%

and clad in SS. The core was made up of a triangular lattice of fuel pins which comprised the highly enriched U alloyed with Be and sintered with BeO to form a ceramic fuel pin. The pins were surrounded by Pb-Bi coolant. The core radius was 390 mm and was surrounded by a SS layer and a BeO reflector, with more Pb-Bi and SS situated above and below the core. The core was penetrated by 10 control and compensation rods (CCRs) and 3 emergency protection rods (EPRs), all constructed of EuB_6 and contained in SS channels. The rods had radii of 10 mm (EPR) and 8.5 mm (CCR). The CCRs were surrounded by approximately 1 mm of Pb-Bi coolant within the SS channel, which was open to the reactor core. The EPR channels were separate from the core and contained no Pb-Bi. In normal operation, Pb-Bi coolant flowed within the CCR channels up to approximately 500 mm above the core. The approximate layout of the core is shown in Fig. 2.

The IASAP corrosion model

Predictions of radionuclide release rates by the IASAP Source Term Working Group required an assessment of the most likely scenarios for corrosion of the SGIs and subsequent water ingress and material release. The development of the corrosion models is described in detail in the IAEA Report (IAEA 1997), and by Warden et al (1997), but it is relevant to give a summary here.

In order to generate the corrosion models, the Group used assumptions chosen to give "realistic worst case" scenarios. The assumptions used were:

- (1) All material of a particular type (e.g. mild steel, stainless steel, Pb-Bi) corroded at a fixed best corrosion rate (BCR), modified by a correction factor. The value of each BCR was chosen to give a fast, but realistic, corrosion rate for conditions in the shallow waters around Novaya Zemlya, using relevant published data on corrosion rates, e.g., Barth and Sheldon (1989), House of Commons Defence Committee (1990). These BCRs, and upper and lower rate values used for sensitivity studies, are summarised in Table 3.
- (2) The correction factors used to modify the BCRs, known as k factors, were dependant on the degree to which the containment barriers had been breached. Values of the k factors lay in the range 0 to 1. They crudely modelled the slowing of the corrosion rate as the oxygen is used inside a volume with little contact to the open sea and the restriction in water flow through the reactor.
- (3) All material was assumed to be released to the environment as soon as it was corroded. This avoided modelling the removal processes from corrosion to the open sea, and immediately provided the worst case.
- (4) The filler materials (Furfurol(F), bitumen and concrete) were assigned a lifetime instead of a corrosion rate. At the time of dumping, the filler was assumed to be a perfect barrier to sea water; the filler material then degraded at a constant rate until, at the end of the assumed lifetime (taken to be 100 years in the primary model), the filler ceased to provide any kind of barrier to water ingress or radionuclide release.
- (5) The presence of fuel pin cladding was ignored as the extent of fuel pin damage was unknown.

The next stage in the modelling process was to ascertain the most probable routes for water ingress to the active material in the dumped objects. In all cases, particular points of weakness such as control rod channels were identified which would lead to water ingress to the SNF. The option of modelling bulk corrosion through the RPV and thermal shields was discounted, since the water ingress at the points of weakness would lead to total corrosion of all SNF prior to any breach of the RPV wall.

Once the route and timing of water ingress were identified, the process of modelling corrosion and release of active material could begin. The BCRs, modified by the relevant k factors, were used to determine the effective rate at which material corroded. The corrosion models hence depended on calculation of the k factors to give the rate at which material was released by

corrosion. For the fuel pins, the rate of material release was also governed by the fuel pin geometry: the greatest release rate would be at the commencement of corrosion, and, as the fuel pin surface area decreased, so would the release rate.

The corrosion model was applied to best Russian estimates of SGI radionuclide inventories (Sivintsev 1993, 1994; Yefimov 1994a) using a FORTRAN code developed at the Royal Naval College, Greenwich, London, and run on a 486DX PC.

Predicted corrosion of SGIs

Applying the IASAP corrosion model and related assumptions to the available information on the structure and material of the dumped SGIs then allowed prediction of likely water ingress routes and corrosion paths.

For the PWRs, the fastest ingress routes into the core were predicted to be via:

- (1) the air bleed tubes used for filling the control rod channels with Furfurol(F), which were sealed with 2.5 mm steel caps;
- (2) the primary circuit inlet and outlet tubes, which had been cropped and welded shut with a 10 mm steel plate; and
- (3) the control rod channels, at the same time as (b), via the 10 mm steel caps on the top of the channels.

Pitting corrosion at 0.5 mm a^{-1} would enable ingress into these routes 20 years after time of dumping. Much later, ingress would be via general corrosion of the RPV structure.

Once water had reached the fuel pins, corrosion would begin at the effective corrosion rate, and material release would occur at a rate dependant on the effective corrosion rate and the area of the fuel pins in contact with water. Radionuclide release would then continue until all active material had gone. For the U-Al alloy fuel assemblages, a faster rate of release was given to some of the more mobile atoms of the fission product inventory, typically 20% of the total activity (Carter 1994). The release of activity from the UO_2 fuel of the icebreaker was modelled by allowing 20% of the total radionuclide inventory to escape from the fuel pins on contact with water, to account for the porosity of the oxide fuel.

The complex construction of the LMRs led to the development of a more detailed model for the corrosion of the LMR cores, based on information supplied by Yefimov (1994a, b). Since the EPR channels were filled with Furfurol(F), it was assumed that, once the bitumen covering the SGI had ceased to be an effective barrier and the control rod caps have corroded sufficiently, water ingress could occur down the EPR channels. The IASAP model calculated this to be at about 125 years after dumping, using a lifetime of 100 years for the bitumen and Furfurol(F) fillers.

Once water had entered the EPR channels, it could corrode the channel SS wall and enter the core. Corrosion was then assumed to progress as shown in Figs. 3 to 5, with water encroaching further into the core until eventually the corrosion sites merged at about the year 24000, leaving the core remnants to corrode until nothing remains at about the year 37000.

Requirements for criticality

The radionuclide release rates predicted by the corrosion model took no account of any change in corrosion rates or fission product inventories due to possible criticality. If a reactor core could achieve criticality, this could potentially have affected the predicted radionuclide release rates in two ways:

- (1) The energy released by fission would cause an increase in temperature in the core, leading to accelerated corrosion rates and hence release rates;

(2) Criticality over a long period would produce a fresh inventory of fission products, some of which would have short half lives not present in the present inventories.

The formulation of the IASAP corrosion models suggested that as corrosion progressed, there was a possibility that some of the corrosion processes could lead to an increase in reactivity of the cores. In particular, the following scenarios were considered:

- (1) Corrosion of a large proportion of the control rod material before the SNF has substantially corroded away. This could have the same effect on core reactivity as control rod withdrawal during a reactor start up, and cause criticality.
- (2) The corrosion process or certain forms of attempted remedial action cause a structural change within the core, such as the SNF falling to the bottom of the RPV or control rods being displaced, resulting in some or all of the SNF and core material reaching criticality.
- (3) Ingress of water into the core causing an increase in neutron moderation (reduction of energy) sufficient to cause criticality.

All the submarine SGIs in which SNF remained still contained a substantial amount of unused ^{235}U : inspection of Table 2 shows that none of the submarine PWRs had used more than about 3.5 kg. Hence a typical dumped SGI fuelled PWR core could be assumed to contain about 47 kg of fissile material, or 94% of the original (start of life) fuel load. The right board LMR in submarine factory number 601 was undamaged and so could be assumed to contain around 89 kg of ^{235}U ; the left board core lost 20% of its load as a result of the accident, so was assumed to contain 71 kg. This suggested the possibility that if the reactivity increased sufficiently by one of the means above, that criticality could occur.

There is no doubt that at the time of dumping the cores retained sufficient U in the correct configuration to form a critical assembly: the reason that they have been dumped is because they either suffered an uncontrolled criticality or had an accident while critical. However, in all cases the reactivity of the core was substantially lowered by the presence of control rods fully inserted and, for the PWRs, filler material in place of water. The problem of criticality thus hinged on two competing reactivity effects: corrosion and subsequent removal of the SNF would decrease reactivity; corrosion of the control rods and other neutron absorbing material, and ingress of water, would increase reactivity.

Criticality calculations are notoriously involved; the probability of criticality for a given amount of ^{235}U depends on a complex relationship of composition, size and shape of the fuel and other core materials, and can vary from a few kilogrammes to many hundreds, depending on the reactor type. As examples (Soodak 1962), the critical mass of a highly enriched sphere of uranium surrounded by a 3 inch BeO reflector is about 17 kg; the critical mass of ^{235}U in 5% enriched 0.6-inch diameter U rods arranged in a hexagonal lattice in water is less than 2 kg.

The design of marine reactors is particularly complex: they have to be able to respond quickly to power changes, and cannot be refuelled during operation. Rapid power changes lead to wide variations in the rates of production of fission products of which some, called reactor poisons, strongly absorb neutrons and thus alter how close the reactor is to criticality. To compensate for short term poison inventories and allow for a useful core life, the core must have a large amount of fuel at the start of life. A high start of life fuel load is achieved in the design of marine reactor cores by using enriched fuel; this fuel load is invariably far more than is required for criticality and, owing to design constraints, cannot always be controlled by the control rods alone. Thus usually the design also includes extra neutron absorbing material such as Gd or B in the fuel assembly (Eriksen 1990). Hence the PWR start of life fuel load of 50 kg (Table 2) would have been far more than the minimum amount required for criticality.

With the limited amount of structural information available to the IASAP, accurate criticality calculations of the dumped cores were not possible; instead some assumptions had to be made. The refuel interval is primarily governed by the amount of useable fuel in the core: too little U and the core cannot be taken critical. Thus the amount of fuel left at refuel gives a good indication of the minimum amount. This can be estimated by examination of Table 2: the core of

unit number 285 achieved a burnup of 2.73 GWd in the 3 years prior to refuelling, giving a rough average of 1 year per GWd burnup. This equates to 1.25 kg of ^{235}U used per year of reactor operation, and suggest that the PWR in unit 285 was inoperable once around 5 kg of U had been used, leaving 45 kg in the core. This figure must allow for a certain amount of control rod material, neutron absorber and poison to be present in the core, all of which significantly increase the amount of U required. Without these, the amount could be much lower, possibly around 30 kg. It should be stressed that the 30 kg figure is a rough working estimate, and is not presented as an accurate amount. On the other hand, the examples given by Soodak above demonstrate that 30 kg is far more than is required in a perfect configuration. Less is known about the fuel requirements of a LMR; since the fuel enrichment and load are much greater than in a PWR, it is likely that the end of life will be at a higher fuel mass, but the 30 kg requirement will be used in this paper. Hence, for all core types, in the following analysis a mass of fuel of less than 30 kg will be considered to be unable to achieve criticality, while a mass of more than that may achieve it.

The next step was to estimate how much control rod material had to be removed before a core would achieve criticality. During normal operation, the core reactivity would have been controlled primarily by the raising or lowering of the control rods to vary the amount of rod material in the core. Shutting down the reactor, either normally or after an accident, would have been accomplished by inserting the rods fully. Raising the rods partially out would start up the reactor: the critical state would equate to a particular rod height. Thus corrosion and removal of the rods would cause a similar effect to raising the rods, and potentially allow reactor start up. During normal operation, the reactors would probably have been operated with the EPRs fully raised to allow safe emergency shut down of the reactor and some of the CCRs at a height which maintained the critical condition. This operating rod height must have been able to vary by as much as possible to allow for poison changes; the optimum rod height for the control rods would hence have been about half core height, or 50% withdrawn. In practice the CCRs may have been operated in groups at different heights to alter the neutron flux profile across the core and compensate for fuel burnup and long term poison changes. There is no information available on submarine PWR rod layout, but the LMRs had 3 EPRs and 10 CCRs. A typical operating configuration of the LMRs would have allowed criticality with ten CCRs at around 50% withdrawal and the 3 EPRs fully withdrawn. This equates to 38% of the total mass of rod absorber in the core, or 62% removed, as illustrated in Fig 6. As with the minimum amount of fuel required for criticality, this figure of 62% should be treated as a rough working estimate only.

Control rod corrosion

All the corrosion models studied by the IASAP use the control rod channel as one of the primary ingress routes for sea water to the core; thus the control rods are subject to corrosion by water as soon as water ingress occurs. For this scenario to allow criticality, the effective corrosion rates of the SNF and the rods must be such that at least 62% of rod material can corrode away before the fuel mass becomes too low to allow criticality, i.e. below 30 kg. The rods and SNF are assumed to be subject to the same containment and filler k factors, so comparison of the control rod and SNF BCRs will be used in the following analysis. However it should be noted that the time values obtained will be incorrect, as the effective corrosion rate will be two orders of magnitude slower than the BCR owing to the containment k factors.

In the case of the submarine PWRs, all control rods were assumed to be constructed of SS, enriched with neutron absorbing material. The BCR of the U-Al alloy is 0.03 mm a^{-1} . If, as discussed above, it is assumed that 47 kg (ie 94%) of fuel was left at the time of disposal, then 30 kg of fuel will be left when the fuel pins radius has decreased from 5 mm to 4 mm. This will take 33 years at the BCR of U-Al alloy (or 1,100 years taking into account the k factors). As discussed above, during normal operation about 38% of the total rod material could be inserted in the core; hence for criticality to occur, about 62% or more of the control rod material must corrode within 33 years. The control rods have a radius of about 10 mm, so to allow 62% to corrode requires a maximum control rod radius after 33 years of 6.2 mm and hence a minimum corrosion rate of $3,8/33 = 0.11 \text{ mm a}^{-1}$, which is not much more than the maximum accepted IASAP SS corrosion rate of 0.1 mm a^{-1} . However, using the minimum IASAP value for the U-Al corrosion rate of 0.015 mm a^{-1} , the assumed minimum critical mass of 30kg will be left after 44

years. The SS corrosion rate required to allow 62% of the rod material to corrode in 44 years is then $3.9/44 = 0.08 \text{ mm a}^{-1}$ which is within the IASAP range of SS corrosion rates. This example suggests that criticality by corrosion of control rods alone is possible if the IASAP corrosion models are accepted as realistic.

The icebreaker PWR used UO_2 fuel with a BCR of 0.0011 mm a^{-1} . However, Container C contained only 60% or 20.6 kg of the total fuel from the N2 reactor which is assumed here to be too low an amount for a critical assembly to form.

The LMR reactors had highly enriched fuel and low burnup. Of all the dumped cores, they contain by far the greatest amount of SNF. However, the ten CCRs were encased in Pb-Bi for 500 mm above the core; the Pb-Bi fills the CCR channels and water can only corrode it from above. Hence the water would have to corrode through approximately 500 mm of Pb-Bi before attacking the EuB_6 in the CCR channels, which would take at least 50,000 years at the BCR for Pb-Bi given in Table 3. Using the IASAP assumptions, the 3 EuB_6 EPRs will begin to corrode after 125 years when water penetrates the elliptical shield, bitumen and EPR channels, and will have corroded completely in around 1000 years. However, corrosion of the 3 EPRs alone is insufficient to cause criticality, as the 10 CCRs will still be fully inserted in the core. As discussed above, the IASAP corrosion model of the LMRs predicts that water will corrode outwards from the EPR channels, removing SNF and Pb-Bi coolant. Eventually the water will reach the CCRs, which are about 160 mm to 173 mm from the EPRs. This will occur at around 17,000 years after dumping, after which the rods will corrode in about 1,000 years. The amount of fuel remaining after 18,000 years can be approximated by taking the ratio of the area of the expanding circles to the total area of the core, as shown in Figure 4; this ratio is around 0.5, i.e. the corroded fuel represents about half of the total initial amount. Hence at the stage when the CCR material corrodes away, about half the fuel is left in the right board reactor, and about 40% in the left, since approximately 20% of the SNF was transported to the steam generator in the accident. This equates to 44 kg of fuel in the right board core, and 35 kg in the left. Hence the IASAP corrosion model suggests that around 18,000 years after dumping sufficient ^{235}U could remain in the cores to form a critical assembly.

Structural changes

Corrosion of SS supporting structure

Alterations in core structure caused by corrosion are, by their very nature, complex and difficult to quantify. An example of a scenario which may lead to criticality would be disintegration of the Furfuro(F) filler after its assumed lifetime and subsequent corrosion of the SS structure holding the SNF, causing the fuel pins to fall into the bottom of the RPV. If sufficient U were left in the fuel pins, and the configuration of the fallen pins formed a suitable arrangement in a small enough volume, a critical assembly could result. The possibility of this would also depend on the amount and arrangement of other materials such as water and corroded control rod material, and would be extremely difficult to model accurately. The rest of this section attempts to determine whether enough U could arrive in the bottom of the RPV to allow the possibility of criticality, but does not attempt to estimate the probability of the U forming a critical assembly. There is little information available about the configuration and thickness of the SS supporting structures; an estimated thickness of 3 mm will be used in this analysis.

Again an assumption is made that 47 kg of ^{235}U remains in the core. In order to leave 30 kg in the pins to provide a critical mass, the pins must only have lost 17 kg of SNF. This will occur if the fuel pin radius has decreased to 4 mm when the SS structure corrodes away.

However, this does not take into account the SNF already corroded. Although the IASAP models assume that all corrosion products are released to the environment, and so provide the most pessimistic release rates, in practice a large fraction of the SNF is likely to be insoluble and remain in the RPV, collecting at the bottom as loose material. The IASAP corrosion model assumed 20% of the fission products in the alloy fuel were more soluble, leaving 80% effectively insoluble. If 80% of the U is assumed to collect in this manner, then an additional amount is available to provide the critical mass. If the IASAP submarine PWR Greenwich FORTRAN model

is run with the SS corrosion rate set at the maximum value in Table 3, and the U-Al alloy rate set at the minimum, the 3-mm thick SS structure corrodes away by about the year 2700. By then the fuel pin radius has decreased from 5 mm to 3.2 mm, losing 60% of the mass of the fuel pin. This could still provide well over 30 kg of SNF at the bottom of the RPV, as shown in the following calculation:

- (1) 60% of the mass of the fuel pins has corroded - 80% of this is insoluble, so $60\% \times 80\% = 48\%$ is lying loose on the bottom of the RPV;
- (2) 40% remains in the fuel pins, which fall to the bottom, giving a total of 88% of the mass of the fuel pins at the bottom, and
- (3) 47 kg or 94% of the SNF is not burnt up, so $94\% \times 88\% = 83\%$ of the start of life fuel load is left on the bottom, which gives a mass of 41.5 kg.

It should be noted that even without the failure of the SS supporting structure the corroded U could fall to the bottom of the core, where it could collect in a compact mass which may be less affected by the presence of neutron absorbers such as the control rods. Hence, assuming the corroded U is able to fall past the remaining core structure and the remnants of the Furfurol(F), the possibility of a critical mass being formed from corroded SNF cannot be ruled out.

This analysis does not take into account any cladding on the SNF, which will increase the probability of enough U remaining in two ways:

- (1) the cladding will slow the corrosion of the SNF, so more will remain once the SS structure corrodes;
- (2) the structural integrity of the fuel pins may depend on the cladding: if the cladding is corroded, the SNF in the form of pellets or small pins may fall to the bottom. The cladding is unlikely to be more than 1-mm thick, so will corrode away before the SS structure, and hence could allow the SNF to fall to the bottom sooner, where more SNF remains.

The presence of neutron absorbing material will increase the critical mass. In the worst case, however, such material may corrode rapidly and cease to affect the core in the time estimated for the rods to corrode.

External corrosion

Corrosion to the external parts of the RPV that weakens its structure has the potential to cause the whole assembly to collapse or topple. The consequences of such an occurrence are difficult to predict. In the worst case, the control rods could be displaced out of the SNF and a critical mass achieved, possibly fast enough to cause prompt criticality (a situation where the increase in reactivity is sudden enough to produce an uncontrolled rise in the fission rate and a rapid, possibly explosive, release of energy). Submarine hull and SGI supporting structures are heavy gauge mild steel, and at an assumed thickness of 25 mm and BCR of 0.08 mm a^{-1} (taking into account the effect of biofouling and corrosion from both sides) could fail after 300 years. The probability of the RPV toppling and the control rods coming out of the core will depend on the configuration of the other structures within the reactor compartment and core.

Any proposed remedial actions which involve lifting or moving the RPV will have to take into account the possibility of the resulting structural change causing criticality: the most catastrophic scenario would be the RPV turning over during transport to shore and the control rods falling out of the core under gravity.

Ingress of water

In the case of the submarine PWRs and icebreaker, the SNF is initially surrounded with Furfurol(F), which was assumed to degrade in 100 years, allowing water to enter the core and

surround the fuel pins. This will increase the reactivity of the core; however, in normal operation the control rods will have been designed to produce a safe shut down condition, and with all rods present there is no risk of criticality. Combined with control rod corrosion and compaction of SNF under its own weight at the bottom of the RPV, however, the presence of water will increase the probability of criticality.

The LMRs were never designed to operate with a water moderator: the reactors in operation were intermediate (ie unlike in a PWR, the neutrons did not have to be fully slowed down to thermal energies to perpetuate the chain reaction), with Be acting as the neutron moderator. If the Pb-Bi were to be replaced by water, a substantial increase in reactivity would result as the higher slowing down power (a measure of the efficiency with which a material slows down neutrons) of the water would produce a much larger thermal neutron flux and hence greater fission rate in the highly enriched U. Giltsov et al (1992) stated that "... in the case of intermediate reactors, one glass of water alone is enough to start a reaction."; this is probably a gross exaggeration, but the presence of water in a reactor designed for intermediate operation is generally thought undesirable. To allow water ingress, however, the Pb-Bi must corrode. This was assumed to occur at the IASAP BCR of 0.01 mm a⁻¹, ten times the rate of the SNF corrosion, so corrosion would remove the Pb-Bi preferentially and replace it with water. The above discussion of control rod corrosion concluded that around 50% of the fuel may be left once water has corroded away all control rod material after about 18,000 years. There are other factors which this analysis did not take into account:

- (1) the preferential corrosion of the Pb-Bi, so more fuel pins will be left than the 50% estimated above;
- (2) the presence of water will further increase the reactivity, leading to less fuel being required for criticality than was necessary during normal operation, and;
- (3) the Pb-Bi corrosion rate has a fastest IASAP value of 0.01 mm a⁻¹, which would reduce the time to reach the CCRs to 2,700 years.

Hence it is possible that owing to the combination of corrosion of the CCRs, the presence of water, and amount of fuel remaining, that as soon as about the year 5000 the reactor cores in submarine factory number 601 could slowly achieve criticality.

The IASAP radionuclide release model also assumes that because the Pb-Bi mixture has a negative coefficient of thermal expansivity, the volume of Pb-Bi has not decreased and remains in close contact with the SNF and SS structures, preventing water from seeping through any gaps and coming in contact with the SNF. Prior to dumping there appeared to be no gaps (Yefimov 1994b). However, the LMRs were dumped in cold Arctic water, and further thermal contraction may have occurred. If the Pb-Bi volume has reduced in comparison to the SS RPV, making water ingress in this manner possible, each fuel pin could eventually be surrounded with a layer of water which could cause a larger increase in reactivity. This would then allow other methods of reactivity increase to occur:

- (1) corrosion of the Pb-Bi layer around the CCRs leading to earlier corrosion of the CCRs;
- (2) corrosion of the SS supporting structures causing the SNF to fall into a smaller volume at the bottom of the RPV, and;
- (3) corrosion of the SS cladding of the fuel rods could result in the fuel pellets being released. If the Pb-Bi has corroded sufficiently, they could fall to the bottom of the RPV, possibly forming a critical mass.

Suffice it to say, the thermal expansion or contraction of the Pb-Bi has a large impact on the potential for criticality, and the time at which it occurs.

Consequences of criticality

Each of the three scenarios alone is unlikely to cause a critical assembly to be formed; however, they are not mutually exclusive, as, for example, control rod corrosion is caused by water ingress. The combination of two or more ways in which reactivity of the dumped SGI cores can increase hence makes the possibility less remote, particularly for the reactors of submarine factory numbers 421, 901 and 601, which have the most SNF remaining and the higher enrichments.

If criticality is possible and occurs through slow corrosion of the control rods and water ingress, the approach to criticality will be extremely slow so the possibility of prompt criticality and any kind of explosion or structural damage can be entirely ruled out. Instead the onset of criticality will cause a slow rise in fission rate, and probably an increase in SNF temperature. The conditions in the core are likely to be such that the heat generated is easily dissipated, particularly as the rate of reactivity increase is so slow. The temperature is unlikely to rise significantly, but may cause a slight rise in corrosion rates and increased flow through the RPV. Since the cores will be water moderated, any rise in temperature will probably cause a reduction in reactivity and there is a possibility that self regulation could occur: this is a condition (deliberately designed into most PWR reactors to produce stable and safe operation) where the reactor naturally remains critical despite perturbations in reactivity caused by material or temperature effects. As corrosion continues, the critical state is likely to be short lived compared with the total lifetime of corrosion, with further structural corrosion and loss of SNF leading to a reduction in reactivity and eventual subcriticality. Such behaviour is unlikely to have much effect on total release rates, unless the critical state exists for long enough to result in significant production of further fission products.

The probability of prompt criticality through structural change is remote, and even if it occurs is unlikely to produce large amounts of radioactivity compared to the present inventories in the cores; for example, the amount of ^{137}Cs generated in a 10^{18} fission criticality excursion (about the same as the SL-1 accident in the United States) would be 44 MBq, which is small compared to the 1994 Kara Sea inventory of about 47000 TBq.

Conclusion

If the IASAP corrosion model and predictions are taken to be substantially correct, then the possibility of criticality of some of the dumped reactors in the Kara Sea cannot be ruled out. The simple estimates and calculations carried out in this paper in the light of the IASAP calculations suggest that:

- (1) in the dumped fuelled cores, there currently exists sufficient ^{235}U in the correct configuration for criticality, but the cores are held sub-critical by the presence of the control rods and, in the case of the PWRs, lack of a water moderator;
- (2) applying the extremes of the IASAP corrosion rates suggests that corrosion of the control rod material alone could be sufficient to allow criticality;
- (3) a combination of control rod corrosion, structural change and water ingress could lead to criticality of some of the dumped PWR cores, and;
- (4) using the IASAP BCR values, a combination of control rod corrosion and water ingress could lead to criticality of the dumped liquid metal cores of submarine factory number 601.

Prompt criticality is only likely to be possible from a sudden structural change, but cannot be entirely ruled out.

REFERENCES

- Barth, C.H. and Sheldon, R.B. "Corrosion Rates of Structural Materials on the Ocean Floor", General Electric Co., Schenectady, NY, KAPL-4701 (1989)
- Carter, J., "Corrosion of LENIN Reactor Compartment Materials", Department of Nuclear Science and Technology, Royal Naval College, Greenwich, London, Technical Memorandum RNC/NS/TM431 (1994)
- Eriksen, V. O., "Sunken Nuclear Submarines", Norwegian University Press (1990)
- Giltsov, L., Mormoul, N. and Ossipenko, L. "The Dramatic History of Soviet Nuclear Submarines - the Feats, Failures and Hidden Catastrophes of 30 years", Robert Laffont (1992) (in French)
- House of Commons Defence Committee, "Decommissioning of Nuclear Submarines," 7th Report, Session 1988 - 1989, Her Majesty's Stationary Office, London (1990)
- International Atomic Energy Agency, "Predicted Radionuclide Release from Marine Reactors Dumped in the Kara Sea - Report of the Source Term Working Group of the International Arctic Seas Assessment Project (IASAP)" IAEA TECDOC-938, Vienna, Austria, (1997)
- Nilsen, T., Kudrik, I., Nikitin, A., "The Russian Northern Fleet - Sources of Radioactive Contamination" Bellona Report 1:1996 (1996)
- Olgaard, P. L., "Nuclear Ship Accidents - Description and Analyses", Technical University of Denmark (1993).
- Sivintsev, Y., "Study of Nuclides Composition and Characteristics of Fuel in Dumped Submarine Reactors and Atomic Icebreaker LENIN: Part I - Atomic Icebreaker", Russian Research Center "Kurchatov Institute," Moscow, Russia (1993).
- Sivintsev, Y., "Study of Nuclides Composition and Characteristics of Fuel in Dumped Submarine Reactors and Atomic Icebreaker LENIN: Part 2 - Nuclear Submarines", Russian Research Center "Kurchatov Institute," Moscow, Russia (1994).
- Sivintsev, Y., "Description of Shielding Barriers at the Nuclear Reactors Dumped in the Arctic", Russian Research Center, "Kurchatov Institute," Moscow, Russia (1995a)
- Sivintsev, Y., "Additional Information on Dumped Ship Reactors and Spent Nuclear Fuel (SNF)", Russian Research Center "Kurchatov Institute," Moscow, Russia, (1995b).
- Sjoebloom, K-L. and Linsley, G., "IAEA Programmes Relevant to the Radioactive Waste Dumped in the Arctic Seas: Part 1. International Arctic Seas Assessment Project (IASAP)", in *Environmental Radioactivity in the Arctic and Antarctic*, P. Strand and E. Holm, Eds., Scientific Committee of the International Conference on Environmental Radioactivity in the Arctic and Antarctic, Ostersund, Norway (1993) pp 88-92
- Soodak, H., (ed.), *Reactor Handbook Vol III Part A - Physics*, 2nd edn, Interscience Publishers (Wiley) (1962)
- Warden, J.M., Lynn, N.M., Timms, S.J., Mount, M.E., Sivintsev, Y., Yefimov, E., Gussgard, K., Dyer, R., Sjoebloom, K-L., "Potential Radionuclide Release Rates from Marine Reactors Dumped in the Kara Sea", *Science of the Total Environment*, in press (1997)
- Yablokov, A. V., Karasev, V.K., Rummyantsev, V.M., Kokeyev, M.Y., Petrov, O.I., Lystsov, V.N., Yemelyanenko, A.F. and Rubtsov, P.M., "Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation", Office of the President of the Russian Federation, Moscow (1993).

Yefimov, E., "Radionuclides Composition, Characteristics of Shielding Barriers, and Analyses of Weak Points of the Dumped Reactors of Submarine N 601", State Scientific Center of Russian Federation, Institute of Physics and Power Engineering, Obninsk, Russia (1994a).

Yefimov, E., personal communication, IASAP Consultant's meeting, Greenwich (1994b)

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Table 1 - Summary of reactor accidents leading to disposal of reactor cores containing spent nuclear fuel (SNF) (Giltsov et al 1992, Olgaard 1993, Nilsen et al 1996)

Factory Number	Class & Hull number	Remarks
901	Hotel K-19	The submarine was on patrol in the North Atlantic in June 1961. Primary pipework ruptured, leading to a loss of primary coolant and core damage. The crew prevented further core damage and probable meltdown by assembling a core cooling system from the drinking water supply. The RC ¹ was subsequently removed.
285	November K-11	An accident occurred during refuelling in February 1965 which led to an uncontrolled chain reaction and fire. The RC was badly damaged and was cut out of the submarine.
	Lenin	During refuelling in 1966 an operator error led to the centreline core being left without cooling water for some time, leading to partial core melt. The design of the original OK-150 RPV ² is unusual in that there is a coolant pipe underneath the RPV, a design that allowed coolant to drain from the RPV and clearly contributed to the accident. The whole of the centreline core barrel was removed with most of the SNF still in place.
421	Yankee II K-140	An accident occurred in August 1968 which led to overpressurisation of the primary circuit. Core damage was suspected, so the RC was cut out and the right board RPV removed for disposal.
601	Mod November K-27	This was an experimental LMR ³ powered vessel. A secondary to primary leak in the LB ⁴ reactor led to fuel channel blockage and core damage, following which an estimated 20% of the fuel pins were transported to the steam generators.

1. Reactor compartment
2. Reactor pressure vessel
3. Liquid metal reactor
4. Left board

Table 2 - Available information on the steam generating installations with spent nuclear fuel dumped in the Kara Sea (Yablokov et al 1993, Sivintsev 1994)

Reactor			²³⁵ U initial conditions		Steam generating installation details					
Unit factory number	Type	Position	Load (kg)	Enrichment (%)	Startup date	Shutdown date	Disposal Location	Disposal date	Burnup (GWd)	Amount of ²³⁵ U at disposal (kg) ¹
901	PWR ²	LB ³	50	20	1961	1961	Abrosimov Fjord	May 1965	1.71	47.86
901	PWR	RB ⁴	50	20	1961	1961	Abrosimov Fjord	Mar 1965	1.67	47.91
285	PWR	LB	50	7.5	1961	1964	Abrosimov Fjord	Oct 1965	2.73	46.59
Ice-breaker	PWR	CL ⁵	75 ⁶	5	1959	1965	Tsivolka Fjord	Sep 1967	14.2 ⁶	34.35
421	PWR	RB	50	20	1968	1968	NZ Depression	1972	1.25	48.44
601	LMR ⁷	LB	90 ⁶	90	1962	1968	Stepovoy Fjord	1981	0.88 ⁶	71.13 ⁸
601	LMR	RB	90 ⁶	90	1962	1968	Stepovoy Fjord	1981	0.88 ⁶	88.91

1. Using a burnup rate of 1.25 kg ²³⁵U per GWd
2. Pressurised water reactor
3. Left board
4. Right board
5. Centre line
6. Second fuel load
7. Liquid metal reactor
8. 20% of the LB reactor fuel was estimated to have been removed from the core at the time of the accident (Yefimov 1994b).

Table 3. Best corrosion rates and lifetimes used for IASAP SGI corrosion models (IAEA 1997)

Material	Best corrosion rate (mm a ⁻¹)	Lifetime (a)	Sensitivity range (mm a ⁻¹)
Stainless steel (bulk)	0.02 ¹		0.1 - 0.03
Stainless steel (pitting)	0.50		0.25 - 0.75
Mild steel (bulk)	0.075 ¹		0.038 - 0.11
Mild steel (pitting)	0.166		0.33 - 0.08
U-Al alloy	0.03 ²		0.015 - 0.045
UO ₂	0.0011		0.0001 - 0.01
U-Be ceramic	0.001		0.0001 - 0.01
Pb-Bi coolant	0.01		0.001 - 0.1
Bitumen ³		100	50 - 500
Furfurol(F) ³		100	25 - 500
Concrete ³		100	50 - 500

1. For steels, bulk corrosion rates were increased by a factor of 2 to account for the effects of biofouling.
2. For insoluble fission products; rate used for soluble fraction is 0.3 mm a⁻¹.
3. Filler materials were given a lifetime in preference to a corrosion rate.

FIGURES

1. *Kara Sea location of dumped reactors containing spent nuclear fuel. Numbers refer to the submarine factory number of the reactors.*
2. *Plan view of the core and reflector of a liquid metal reactor of submarine factory number 601, showing the approximate position of the emergency protection rods and control or compensation rods.*
3. *Circles of corrosion expanding from the emergency protection rods of a liquid metal reactor of submarine factory number 601.*
4. *Circles of corrosion from emergency protection rods (EPRs) at about 16,400 years after dumping, after stainless steel (SS) thermal shields and BeO reflector outside the core have corroded away. The expanding circles centred on the EPR channels have almost reached the control or compensation rods.*
5. *Circles of corrosion merging with corrosion from the outside of the core at about 18,000 years after dumping. All control rods have corroded away.*
6. *Schematic of a reactor core showing emergency protection rods (EPRs) fully withdrawn and control and compensation rods (CCRs) at a typical operating height of 50%. Only 4 out of 10 CCRs are shown for clarity.*

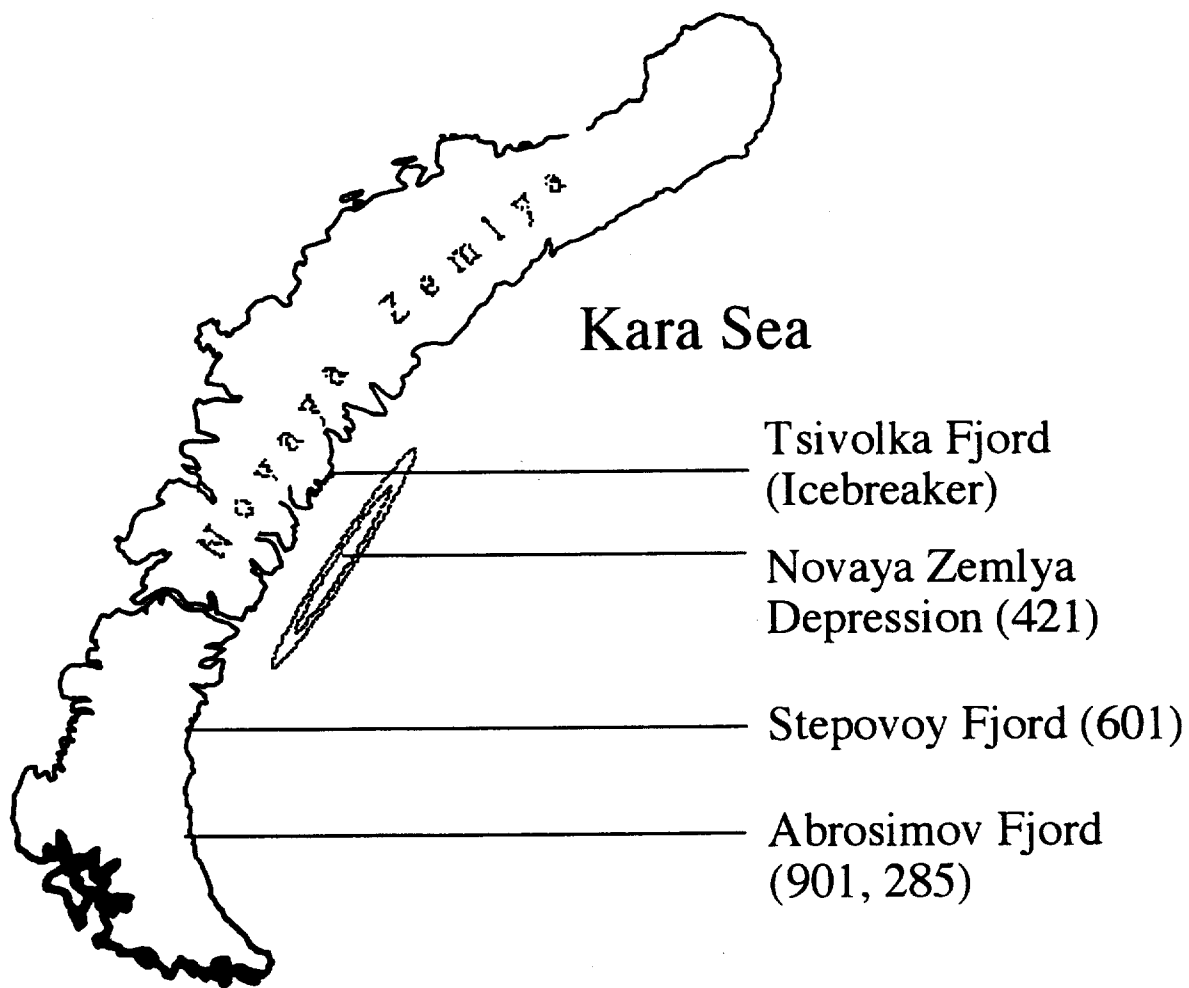


Fig. 1 - Kara Sea location of dumped reactors containing spent nuclear fuel.

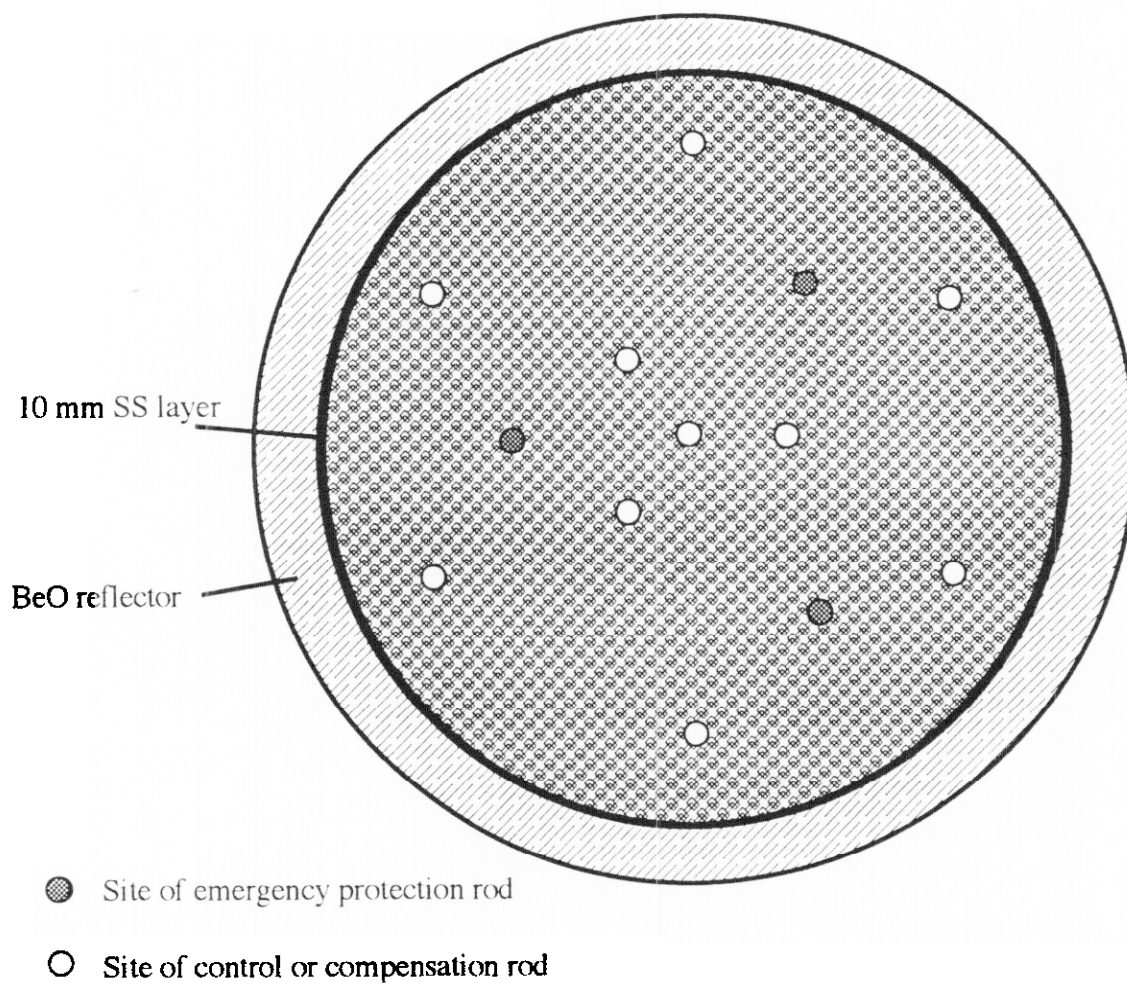


Fig. 2- plan view of the core and reflector of a liquid metal reactor of submarine factory number 601, showing the approximate position of the emergency protection rods and control or compensation rods

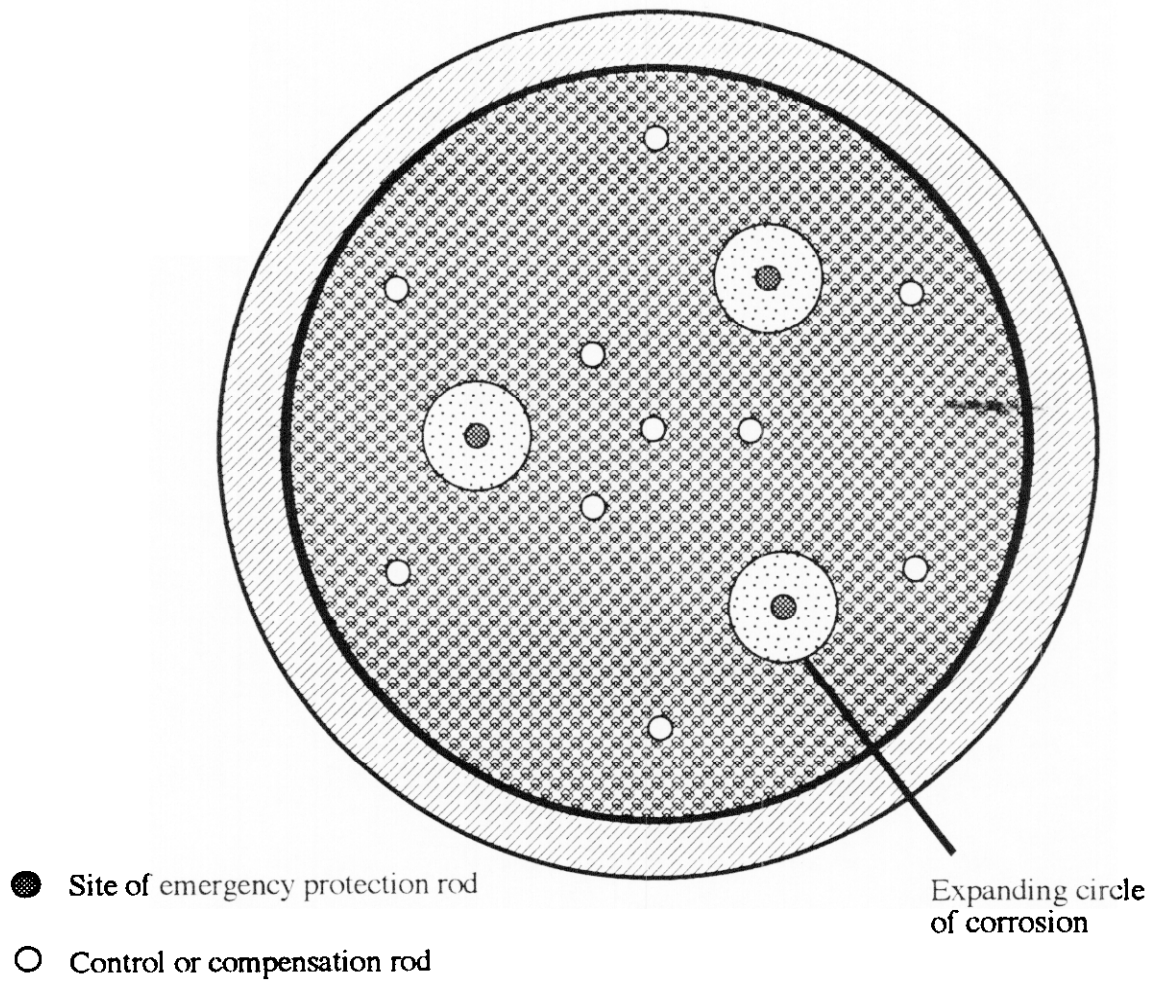


Fig. 3 - Circles of corrosion expanding from the emergency protection rods of a liquid metal reactor of submarine factory number 601.

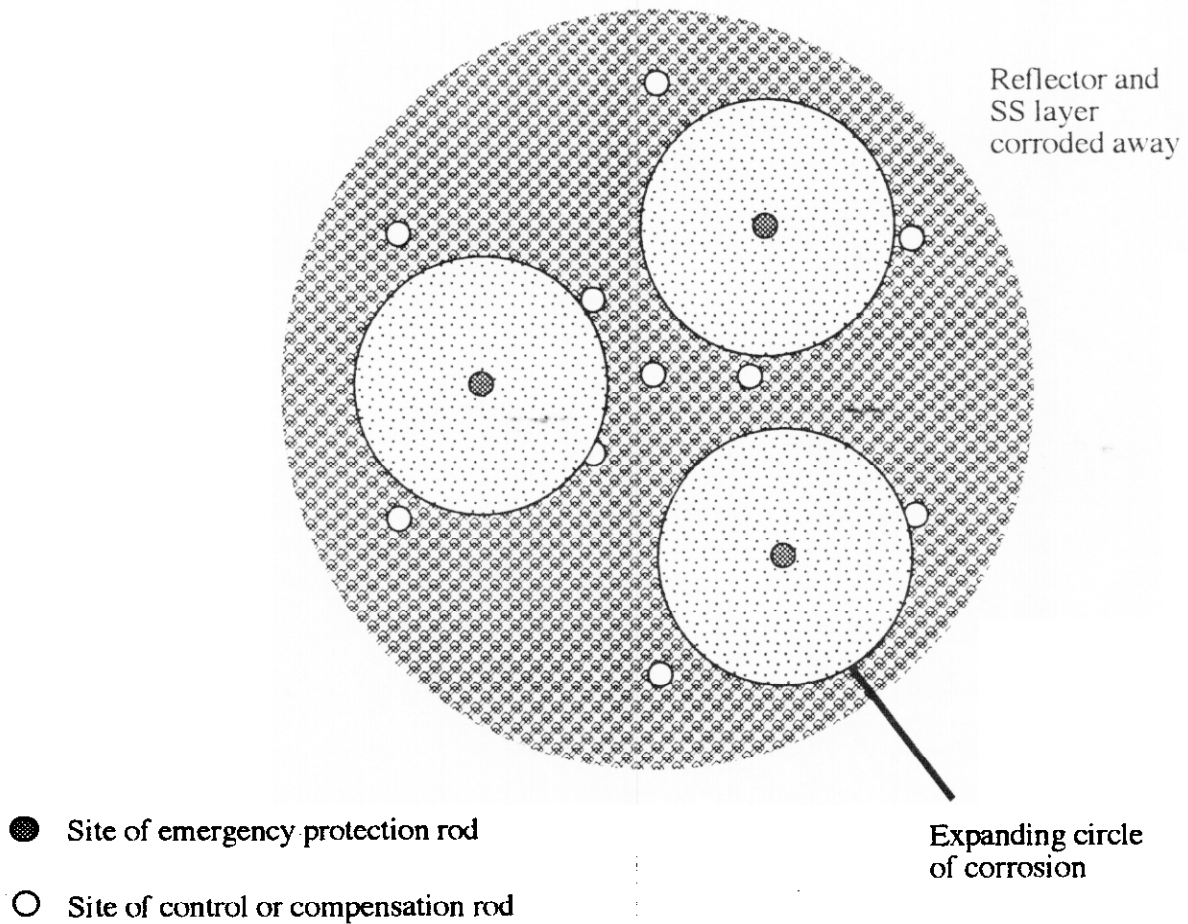


Fig. 4- Circles of corrosion from emergency protection rods (EPRs) at about 16,400 years after dumping, after stainless steel (SS) thermal shields and BeO reflector outside the core have corroded away. The expanding circles centred on the EPR channels have almost reached the control or compensation rods.

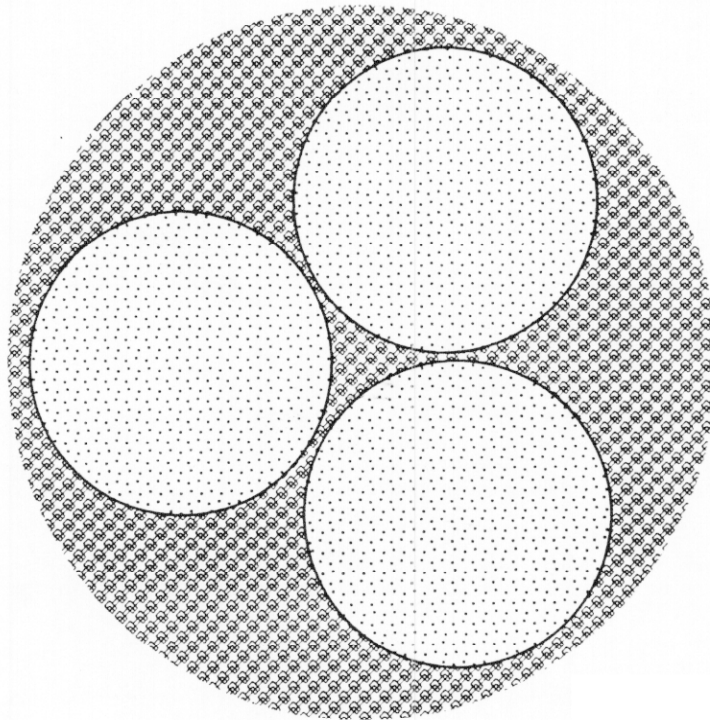


Fig. 5- Circles of corrosion merging with corrosion from the outside of the core at about 18,000 years after dumping. All control rods have corroded away.

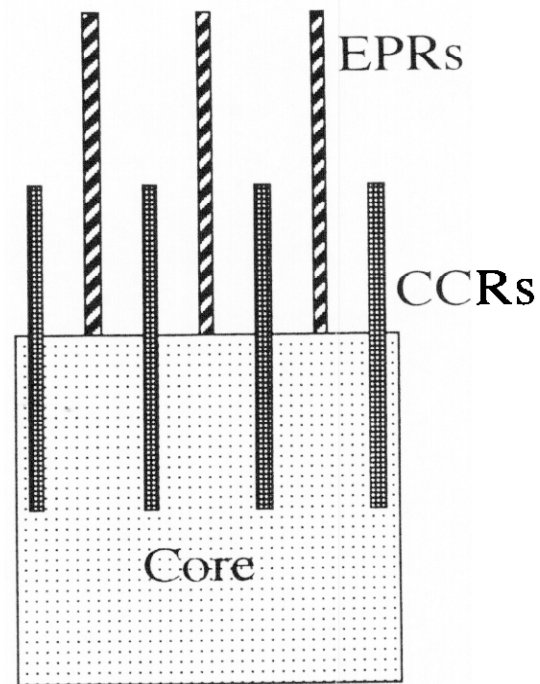


Fig. 6 - Schematic of a reactor core showing emergency protection rods (EPRs) fully withdrawn and control and compensation rods (CCRs) at a typical operating height of 50%. Only 4 out of 10 CCRs are shown for clarity.

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