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WF Brehm

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92. START

CONTROL OF RADIOACTIVE MATERIAL TRANSPORT IN SODIUM-COOLED REACTORS

W. F. Brehm

March 1980

Department of Energy

Environmental Control Symposium

March 17, 1980, Reston, VA

HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company, a subsidiary of Westinghouse Electric Corporation, under the Department of Energy Contract No. EY-76-C-14-2170

CONTROL OF RADIOACTIVE MATERIAL TRANSPORT IN SODIUM-COOLED REACTORS

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INTRODUCTION

The Radioactivity Control Technology (RCT) program was established by the Department of Energy to develop and demonstrate methods to control radionuclide transport to ex-core regions of sodium-cooled reactors. This radioactive material is contained within the reactor heat transport system with any release to the environment well below limits established by regulations. However, maintenance, repair, decontamination, and disposal operations potentially expose plant workers to radiation fields arising from radionuclides transported to primary system components. The development of successful techniques to control radioactive material transport means lower occupational radiation exposure to workers, lower maintenance costs, improved plant factor and reduced doubling time for breeder reactors.

Three distinct classes of radioactive species are released: activated corrosion products from fuel cladding and core components, fission products from breached fuel pins, and tritium. The corrosion products and tritium are always present; fission products are released to the sodium only if fuel pins breach.

In order to be economically viable, sodium-cooled reactors must operate at fairly high temperatures (500°C) and with long, uninterrupted fuel irradiation runs. These conditions increase the potential for radioactive corrosion product transport; achieving the goal of long uninterrupted operating may mean that release and transport of fission products from small numbers of breached fuel pins may have to be controlled. The radioactivity transport problem is not new or unique to sodium-cooled reactors; it is also seen in light-water reactors.

An example of the problems that have been encountered in liquid-metal cooled reactors was seen at the Dounreay Fast Reactor (now decommissioned) in Great Britain, where location and repair of a leak in the coolant circuit lasted several months and required hundreds of workers to limit exposure to allowable levels.([1]) (Furthermore, allowable exposure to people was greater than it is today.) Conversely, reduction of radiation levels has been achieved in the decontamination of the BR-5 reactor in the Soviet Union,([2]) the decontamination prior to repair of the PHENIX heat exchangers in France,([3]) and the removal of ^{137}Cs from sodium in the Experimental Breeder Reactor-II (EBR-II) in the United States.([4])

It is to be emphasized that essentially all the radioactive material remains within the confines of the primary heat transport system. There is no radiation exposure to the public, and exposure to plant workers only when it becomes necessary to perform maintenance and repair operations on or adjacent to primary system components.

the fuel particulates will probably contain significant amounts of fission products, and also because of the need to control transport of Pu in the primary system if the (U,Pu) fuel cycle is used. Evidence to date suggests low solubility of Pu in sodium and low mobility of fuel particulates in reactor primary systems.

TABLE 1. RADIONUCLIDES

Nuclide	Formation Reactions	Half-life Days	Gamma Energy (MeV)	Comments
^{54}Mn	$^{54}Fe(n,p)$+ $^{55}Mn(n,2n)$	313	0.84	Most abundant corrosion product
^{60}Co	$^{59}Co(n,\gamma)$+ $^{60}Ni(n,p)$	1913	1.17, 1.33	Source is Co impurity in nickel, plus Co-base alloys
^{58}Co	$^{58}Ni(n,p)$ $^{59}Co(n,2n)$	71	0.81	Unimportant relative to ^{54}Mn and ^{60}Co
^{59}Fe	$^{58}Fe(n,\gamma)$+	45	1.10, 1.29	
^{137}Cs	fission product	1.1×10^5	0.66	Dominant fission product
$^{140}Ba/La$	fission product	12.8/1/6	0.57	Observed if extended operation with failed fuel
$^{95}Nb/Zr$	fission product	35/65	0.76	
^{131}I	fission product	8	0.36	Unimportant in steady state operation, only accident conditions
^{125}Sb	fission product	996	0.43	
^{239}Pu	fuel	8.9×10^6	5.1 (alpha)	
^{3}H	$^{10}B(n,\alpha)^{7}Li(n,n\alpha)$+ $^{10}B(n,2\alpha)$ ternary fission	4500	0.006 (beta)	
^{65}Zn	$^{64}Zn(n,\gamma)$	274	1.11 0.51	From impurities in sodium and structural metals, seen in Europe but not in the USA.

+dominant reaction

This paper deals with radioactive material generated and transported during steady-state operation, which remains after ^{24}Na decay. Potential release of radioactivity during postulated accident conditions has been treated extensively elsewhere and will not be discussed here. The paper discusses the control methods for radionuclide transport, with emphasis on new information obtained since the last Environmental Control Symposium,([5]) and shows that development of control methods is an achievable goal.

SUMMARY

A large number of corrosion and fission products have been identified in the primary circuit of sodium-cooled reactors. Reducing oxygen level in sodium reduces corrosion product release by a small amount, but not enough to solve the transport problem. Lowering reactor operating temperature will also reduce the corrosion product source rate, but to achieve a complete solution would require temperatures too low for practical power production.

Engineered devices to remove corrosion products have proven quite successful in the first in-reactor tests; several modifications to the original concept which may improve their efficiency and reduce installation cost, are scheduled for test. Removal of soluble fission products (mostly ^{137}Cs) by other engineered devices and cold trapping has been demonstrated. First data from a "deposition sampler" run in conjunction with run-beyond-cladding breach tests are presently being evaluated.

A process for component decontamination, leaving the component qualified for return to service, has been developed for 304 stainless steel, and is being adapted to other materials.

Tritium permeation rates through reactor materials under operating conditions are quite low; furthermore, operation of sodium purification systems (cold traps) at low temperature will precipitate most of the tritium in the cold traps.

Future work will concentrate on demonstration of trapping and decontamination processes, improving transport prediction methods using operating data from reactors, and working on control of radioactive material during fuel handling, storage, and waste disposal and plant decommissioning.

DISCUSSION

Radionuclide Sources. Table 1 lists the radionuclide sources, together with their half-lives and gamma-ray energies. Tritium (^{3}H) has no gamma ray, but, because it is mobile and can be absorbed into water supplies, control of tritium transport is required. The most prevalent nuclides are ^{54}Mn, ^{60}Co, ^{137}Cs, ^{140}Ba-La, ^{95}Nb-Zr, and ^{3}H. Large quantities of ^{24}Na are produced during reactor operation, but it will decay to negligible amounts in one to two weeks after plant shutdown because of its 15-hour half-life. Sodium-22 (^{22}Na), although present, will not be a significant contribution to radiation dose rates, compared to the corrosion and fission products.

Continued operation with breached fuel pins may result in release of fuel particulates to the primary system. This event warrants study because

the fuel particulates will probably contain significant amounts of fission products, and also because of the need to control transport of Pu in the primary system if the (U,Pu) fuel cycle is used. Evidence to date suggests low solubility of Pu in sodium and low mobility of fuel particulates in reactor primary systems.

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^{65}Zn	$^{64}Zn(n,\gamma)$	274	1.11 0.51	From impurities in sodium and structural metals, seen in Europe but not in the USA.

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Information from radionuclide transport measurements in test loops and operating reactors has shown that much of the transported corrosion product radioactivity is adherent to the deposition sites and is not removed by draining away the sodium, or by water or alcohol washes. The most prevalent fission product, ^{137}Cs, does not adhere strongly to surfaces. The ^{137}Cs is usually found in locations where large volumes of sodium remain after draining, also in lower-temperature regions such as cold traps.

Schematic representations of nuclide transport and deposition are shown in Figures 1, 2, and 3. Keeping the radioactive material within the reactor vessel (as can be done with a "nuclide trap" which is an integral part of the fuel assembly) precludes any further interaction with people; precipitation and removal of radioactive material in the purification system also provides isolation. Precipitation of tritium in the secondary system cold trap provides less isolation,* but the tritium itself is safely confined, and the trap material itself provides an effective barrier against the weak 0.006 MeV beta emission from tritium.

Several studies and experiences[3,6,7,8] have shown that radiation levels over 1 R/hr can be expected near reactor primary system components from corrosion product (^{54}Mn, ^{60}Co) release and transfer; addition of fission products from breached fuel has created radiation fields of the same magnitude.[8] Because of the radiation exposure to workers, extended operations in fields of over 200 mR/hr are cumbersome and expensive, and a field of 25 mR/hr is the goal of some modern plant designs. It is to be emphasized that these radiation levels are for workers in the plant, that the goal remains to have negligible release of radioactive material outside the plant; sodium-cooled reactors operating to date have achieved this latter goal.

A multi-phase program has been established to reduce the amount of transported radioactivity and develop a decontamination process for components as shown in Figure 4. The status and recent accomplishments of each control technique are given below.

Coolant Purity and Temperature Effects. Experimental studies were completed showing that reducing oxygen in sodium from 2.5 to 0.5 ppm had little effect on ^{54}Mn release at both 604°C and 538°C, and reduced ^{60}Co release by a factor of two to three. Substantially more ^{54}Mn than ^{60}Co is released because ^{54}Mn is preferentially released from the stainless steel, while ^{60}Co is preferentially retained at the metal surface. The ^{54}Mn release rate at 604°C was three times that at 538°C, the ^{60}Co release rate was 1.7 times as great.[9,10,11] These results contradict the hypothesis of Reference 6, but the observed effect of oxygen on nuclide release was factored into the more recent work of Reference 7. Operations at < 0.01 ppm oxygen with a hot trap at 604°C did not decrease the ^{54}Mn or ^{60}Co release below that observed at 0.5 ppm oxygen.[12] Considerable information was obtained on

*The tritium gets into the secondary system of the reactor by diffusion through the walls of the intermediate heat exchanger, a quite slow process.

nuclide distribution around a sodium loop; the ^{60}Co favors the hot leg as a deposition site while the ^{54}Mn deposits preferentially in the cold leg. Some (probably only 1 or 2 percent) of the released activity is circulated around the loop before depositing. Both ^{54}Mn and ^{60}Co that deposit in the hot leg diffuse into the metal substrate, whereas the ^{54}Mn in the cold leg is found in Ni-Mn rich deposits on the metal surface, adherent to it. The deposits are more adherent at higher temperatures and lower oxygen levels. The conclusions from this study, which is now complete, are that changes in operating temperature and oxygen levels within limits necessary for practical plant operation, will not stop corrosion product radionuclide transport.

Radionuclide Trap Development. Pure nickel was found to selectively remove ^{54}Mn from sodium in the hot leg of a system. Experimental traps were tested in small test loops; from these results a series of full size trap tests for EBR-II and FFTF were developed, using 0.13 mm (5 mil) pure nickel sheet wound on a mandrel, and dimpled to maintain 1.1 mm (45 mil) interlayer spacing.

The rolled sheet is inserted in fuel assemblies directly above the fuel pins. In this position the trap is exposed to all sodium passing through the assembly, and the trap is changed when fuel is changed as shown in Figures 5 and 6. Two tests have been completed in EBR-II. Results of the first test show that the trap takes up 5-9 times as much ^{54}Mn as is released from the surfaces below; therefore the trap is acting as a getter for the circulating ^{54}Mn in the entire primary sodium system; analysis of the second test is nearly complete, and the data generally agree with the first test.

A vapor-blasted surface proved more effective in small loop tests at collecting ^{54}Mn than a conventional surface. A nickel plated surface was equally effective (per unit area) as nickel sheet. This result leads to an alternative trap approach, nickel plating the fission gas plenum region of the fuel pins. Doing so will enable the total core length to stay the same and eliminate most of the extra pressure drop through the fuel assembly (the trap adds some). The combination of rolled-sheet traps in outer row assemblies and nickel plated fuel pins in inner fuel assemblies appears to be promising in reducing ^{54}Mn transport to the ex-vessel piping and components.

Fission Product Transport and Control. Cesium-137 was found to be the most prevalent fission product released when "gas leaker" types of breached fuel occur. The ^{137}Cs is part of a fission gas decay chain. Other fission products, such as ^{140}Ba-La and ^{95}Zr-Nb, are released only by direct recoil or if there is fuel-sodium contact, and in much smaller amounts. The ^{137}Cs is soluble in sodium, non-adherent to surfaces, and deposits preferentially in cold traps. The other fission products are far less soluble in sodium and do not preferentially migrate to cold traps.

Cesium has successfully been removed from sodium by carbon and graphite in loop tests; the entire EBR-II primary system had ^{137}Cs removed while the reactor was shut down using reticulated vitreous carbon (RVC), which has roughly the same porosity and physical characteristics as styrofoam.([4])

A schematic of this trap is shown in Figure 7. This effect has been characterized quantitatively at our laboratory. Three traps using graphite and reticulated vitreous carbon were tested in a small loop at HEDL. ^{137}Cs was successfully removed from the loop with no detectable increase in the carburizing potential in the loop; the RVC and graphite were equally effective at removing ^{137}Cs, but the RVC remained undamaged.

An improved trap design was developed and a test unit was fabricated for testing.

To better understand the release and transport characteristics of fission products and fuel particulates, an experimental program has been set up in EBR-II, which includes a "deposition sampler" in a location above a fuel assembly with a breached fuel pin.([13]) This test is shown schematically in Figure 8. The first test has been completed. Considerable fission product radioactivity, especially ^{95}Zr-Nb, ^{140}Ba-La, ^{144}Ce, ^{103}Ru, was present; ^{137}Cs was absent as expected. Considerable ^{54}Mn was found on the nickel specimens which were included in the sample train. Results from these tests will better define fission product source rates from breached fuel and establish the effectiveness of the nickel (which may be part of a trap for ^{54}Mn as described previously) at removing fission products. These results will be used to establish limits for operation with breached fuel.

Decontamination. Most decontamination process development work to date has been on a hot leg decontamination process in order to be ready should a pump need to be decontaminated. Analysis of deposition and diffusion data showed that removal of 20 μm (0.75 mil) of material would remove most of the radioactivity. A limit of 50 μm (2 mils) maximum intergranular attack from the decontamination process has been established. A solution of 2-1/2% glycolic-2-1/2% citric acid in water at 70-90°C, with dissolved oxygen maintained at < 10 ppb has been found effective for wrought 304 stainless steel.([14])

Reactors will use 304 and 316 stainless steel in both cast and wrought form, for primary system components. The glycolic-citric acid process is being evaluated for application to these materials. A diffusion study to determine whether ^{54}Mn and ^{60}Co diffuse equal distances into 304 and 316 castings and wrought material (these results could affect the 20 μm material removal depth) is underway.

The Ni-Mn rich deposits in the lower temperature section of a reactor intermediate heat exchanger and in reactor cold legs([15]) are not removed by the glycolic-citric acid process used for the hot leg. A different process is being developed.

Radioactivity Transport Analysis. Calculational methods have been developed for estimating the radiation levels in a reactor from corrosion product transport. The model is improved as new data are obtained.

One inch diameter surveillance holes have been cast into the top of one of the FFTF heat transport system cells. When the plugs are removed from the holes, a well-collimated bean of gamma rays will be available for analysis. The required detector and multichannel analyzer have been procured, set up and calibrated. This is shown schematically in Figure 9.

Tritium Control. An experimental program investigated the permeation of tritium through reactor containment materials, and tritium control methods. It was found that permeation through 304 and 316 stainless steels and Fe-2 1/4 Cr-Mo ferritic steels was much slower than anticipated because of oxide layer formation on the non-sodium-wetted side of the material. Maintaining a cold trap temperature as low as possible (~ 115°C) reduced tritium activity in the sodium to a very low level. Since co-precipitation with sodium hydride assists tritium precipitation, hydrogen entering a reactor secondary system from the steam generator circuit will also aid in keeping tritium activity in the secondary system quite low. The conclusion of the work was that tritium release from a sodium-cooled reactor during normal operation is not a problem.([16])

In summary, we believe that trapping and decontamination give the possibility to restrict and control radionuclide transport and minimize radiation exposure to plant workers.

Future Work. Work in progress in the Radioactivity Control Technology program will concentrate on reactor demonstration of the traps for ^{54}Mn and ^{137}Cs, and development of the improved ^{137}Cs trap. Reactor operating data on corrosion and fission product transport will be obtained; fission product release during operation with breached fuel will be obtained from the tests described previously. Development and demonstration of decontamination process compatible with all reactor materials will be completed. Results of these test programs will be factored into the Conceptual Design Study (CDS) for a large sodium-cooled reactor.

Related Studies. There are other closely related programs that deal with control of radioactive materials. For example, an effort is underway to develop a process to remove most radionuclides from waste sodium generated during component replacement and repair, then convert the remaining sodium to glass for safe disposal (or alternately reserve the sodium for re-use in a reactor). Quantities (several hundred kilograms) of sodium and sodium components containing tritium, sodium-22, and corrosion and fission products will have to be processed when purification cold traps are replaced. An evaporative process for removal of sodium from fuel assemblies is being developed, this process will be most useful to remove sodium from breached fuel assemblies and avoid energetic reactions between water and highly irradiated fuel. Large quantities of mildly acid or alkaline water containing low concentrations of radioactivity will be generated during sodium removal and component decontamination operations; the technology already exists to concentrate the radioactivity for safe disposal.

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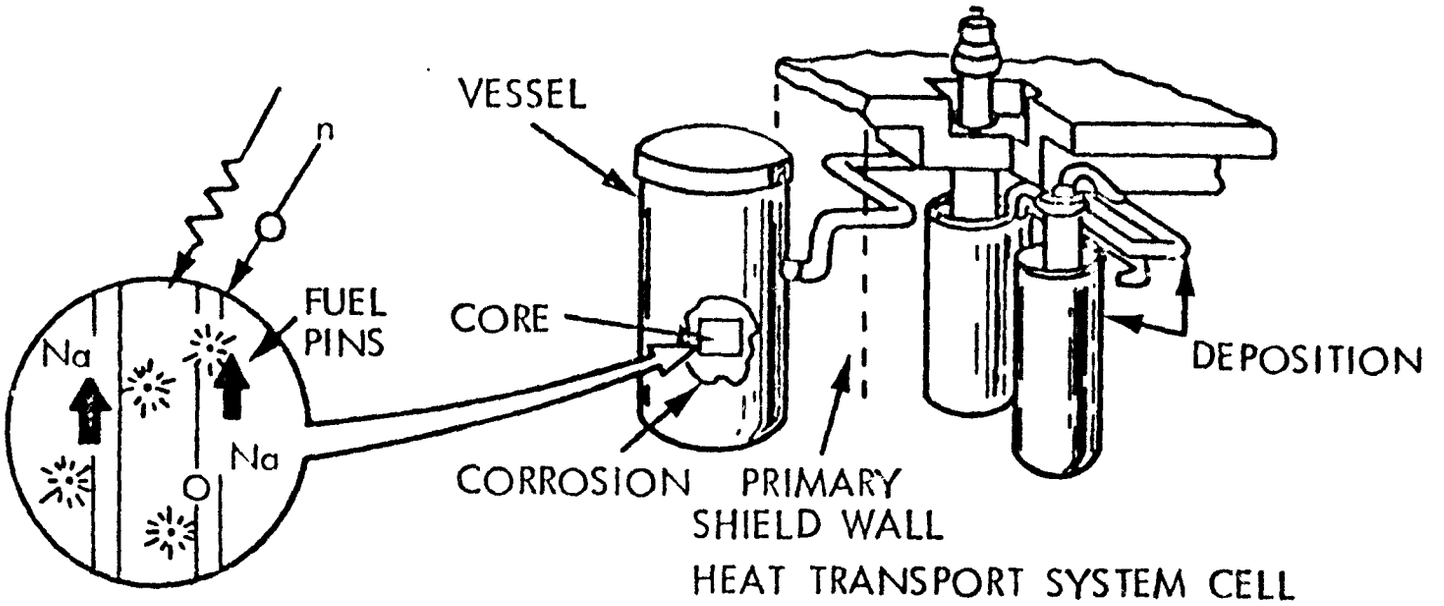


ACTIVATED CORROSION PRODUCTS ^{54}Mn

^{60}Co

^{58}Co AND OTHERS

ACTIVATED CORROSION PRODUCT TRANSPORT

FIGURE 1

HEDL 7809-118.6

FISSION PRODUCT RELEASE

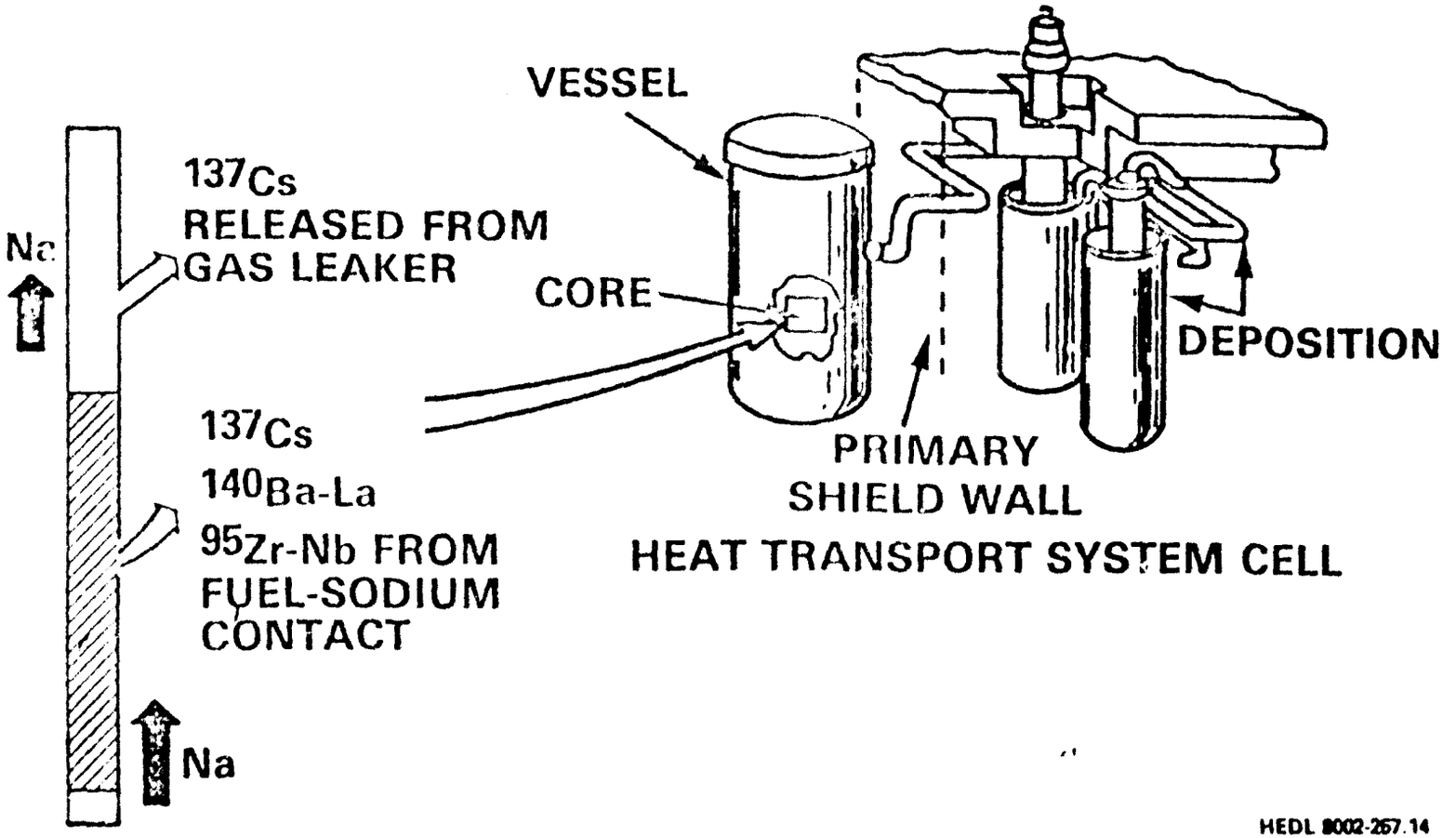


FIGURE 2

LOOP-TYPE REACTOR SCHEMATIC

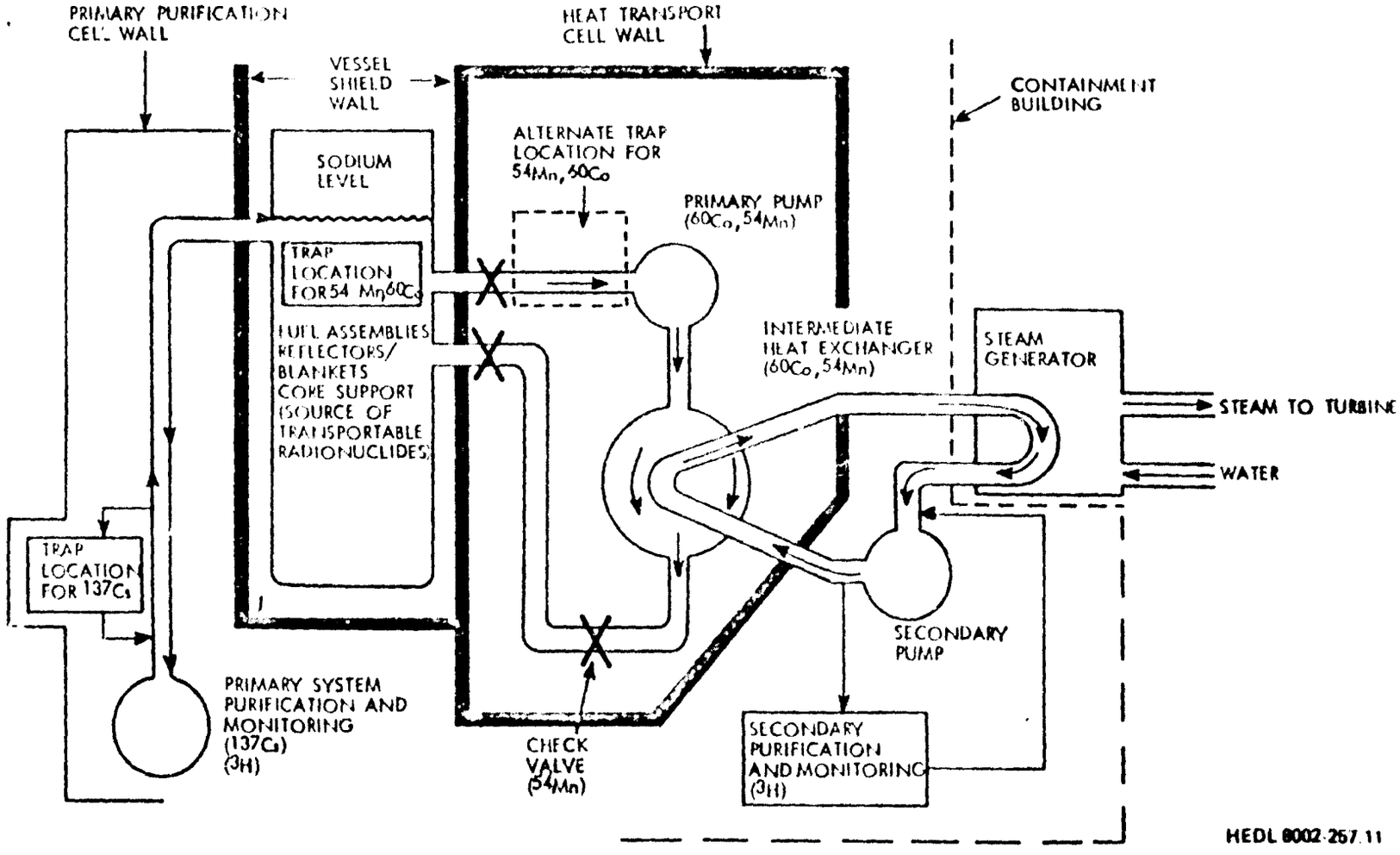
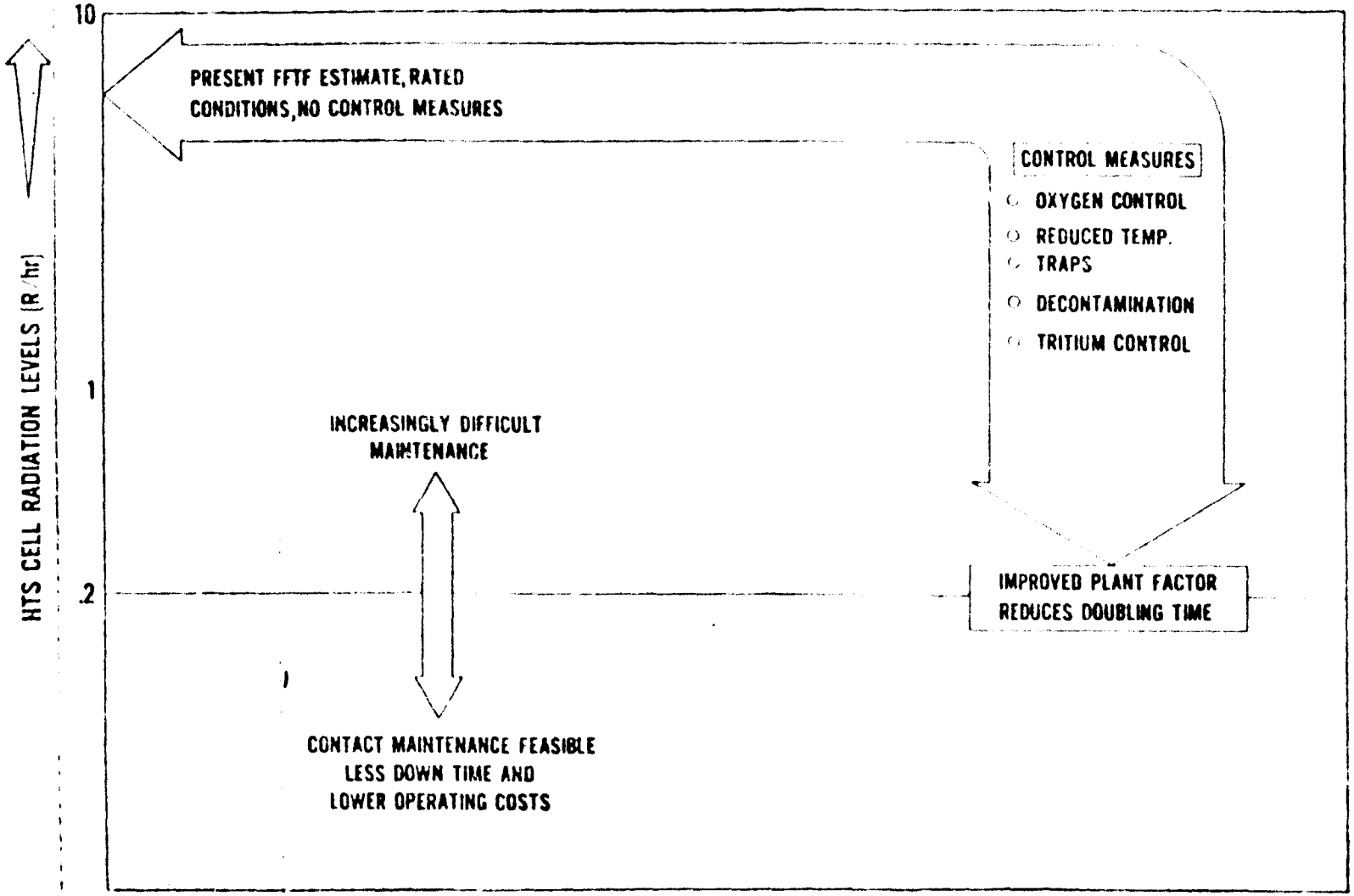


FIGURE 3

CORROSION PRODUCT TRANSPORT CAN CAUSE MAINTENANCE PROBLEMS

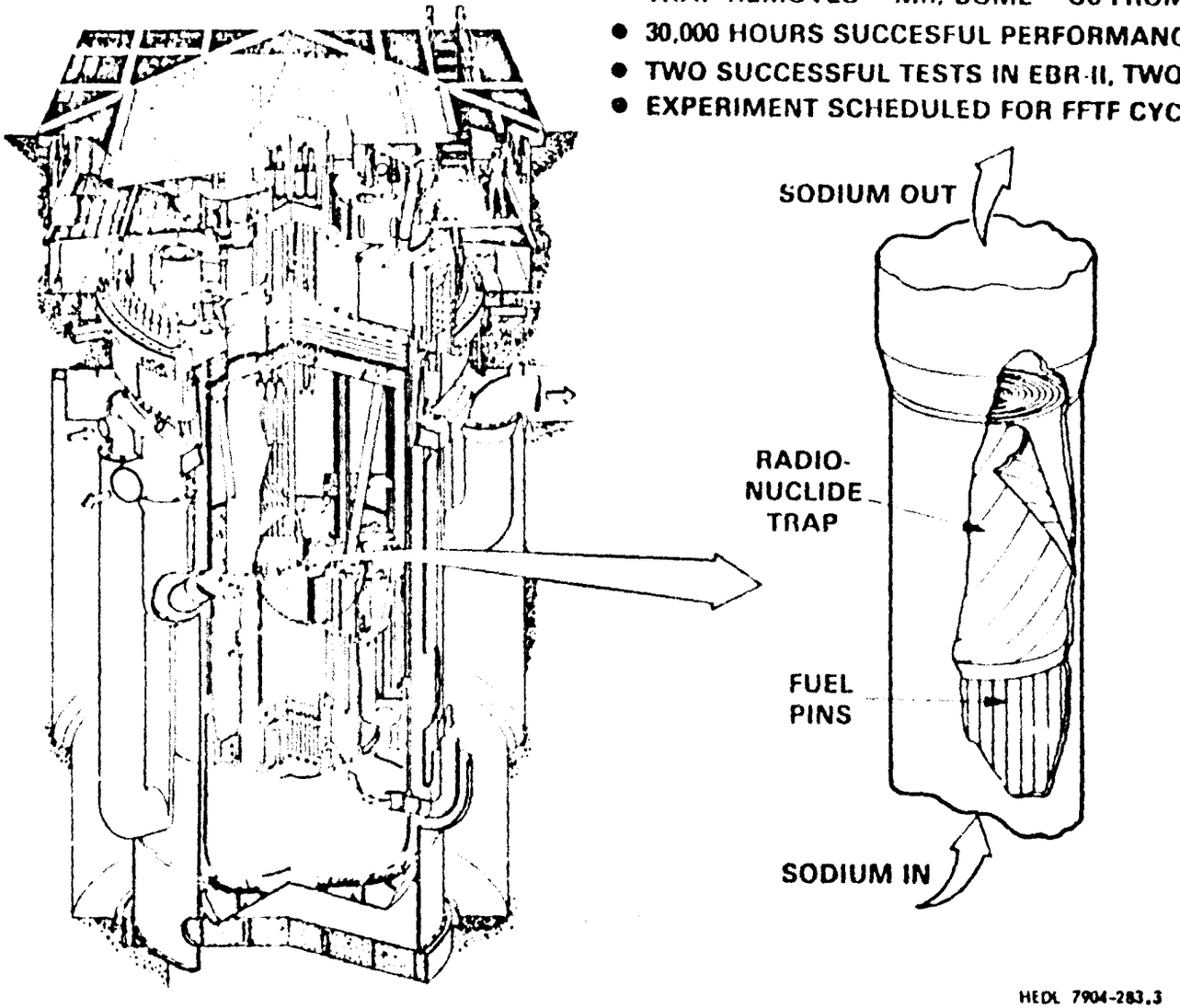


HEDL 7612-142.2

FIGURE 4

RADIONUCLIDE TRAP

- TRAP REMOVES ^{54}Mn, SOME ^{60}Co FROM SODIUM
- 30,000 HOURS SUCCESFUL PERFORMANCE IN TEST RIGS
- TWO SUCCESSFUL TESTS IN EBR-II, TWO MORE GOING IN
- EXPERIMENT SCHEDULED FOR FFTF CYCLE 1



HEDL 7904-283.3

FIGURE 5

FTR RADIONUCLIDE TRAP

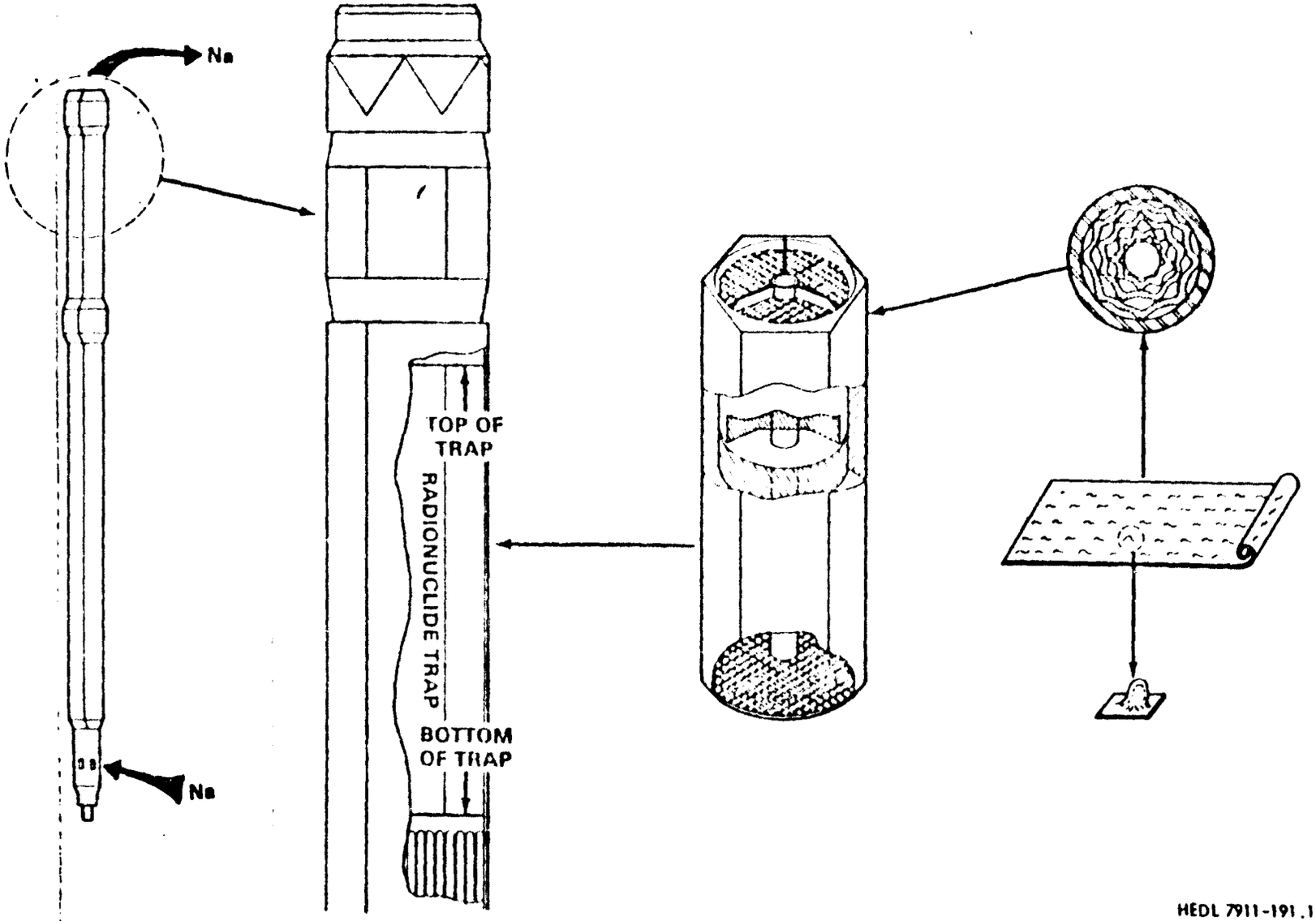


FIGURE 6

EBR-II CESIUM TRAP

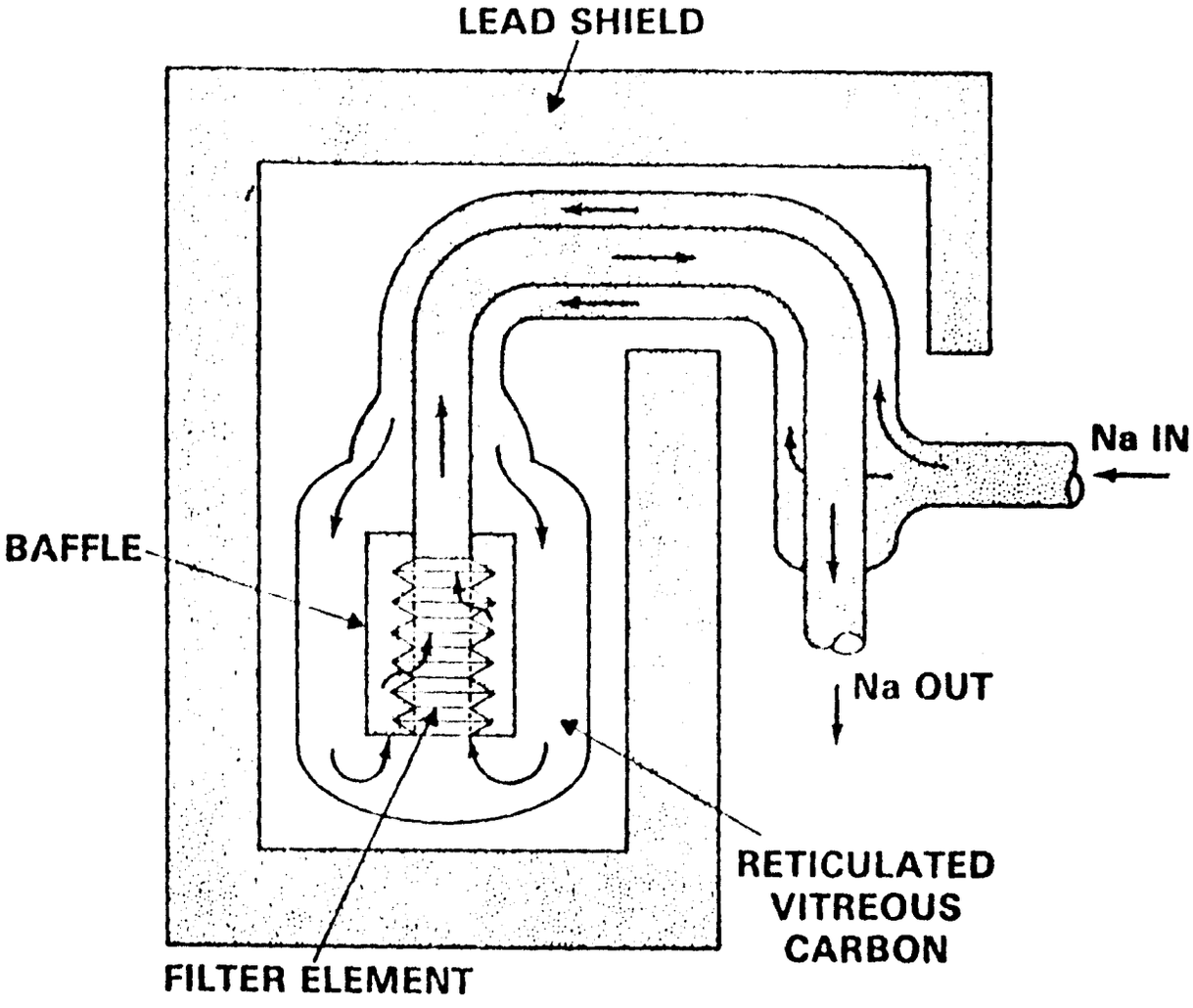


FIGURE 7

DEPOSITION SAMPLER (SCHEMATIC)

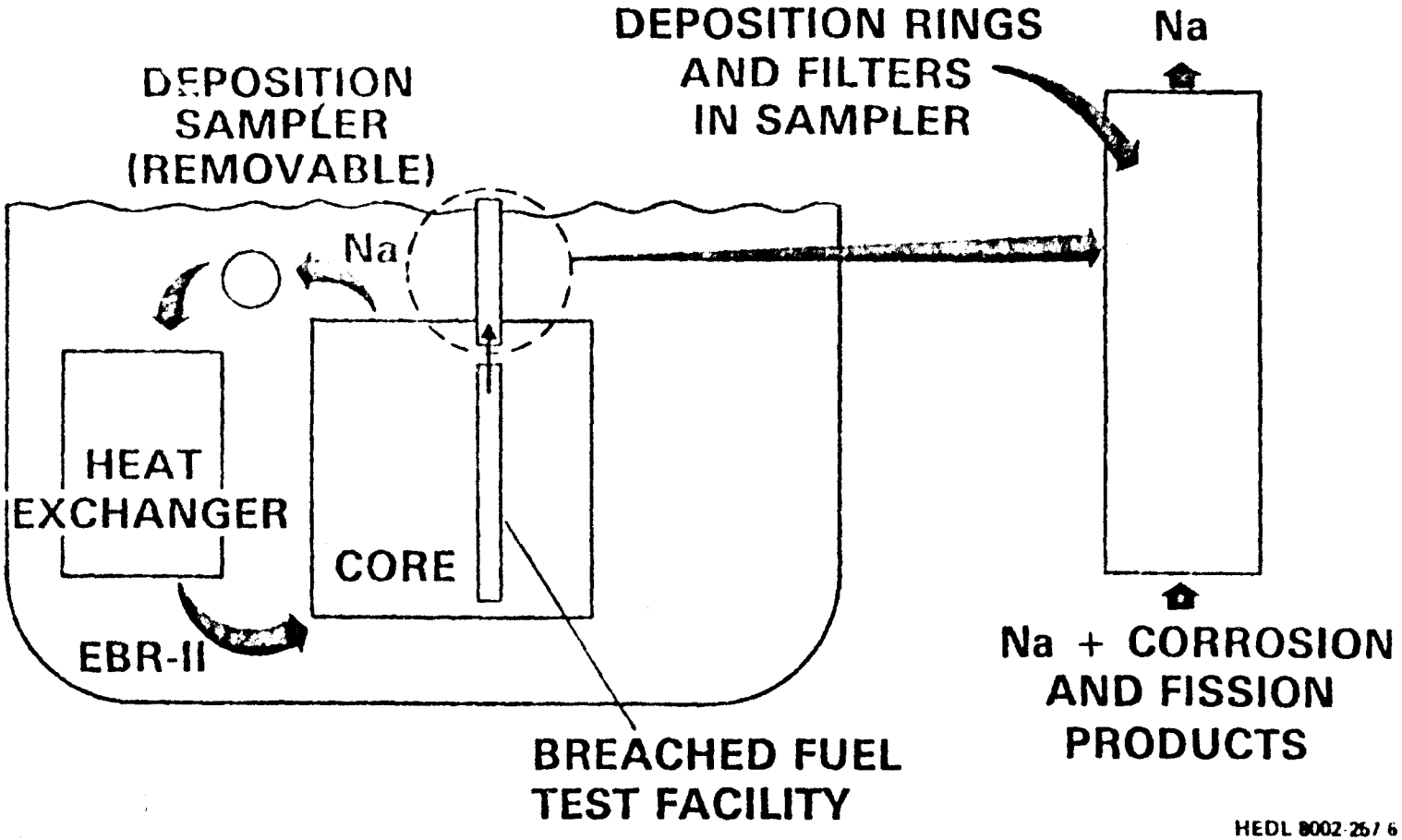


FIGURE 8

RADIOACTIVITY BUILDUP SURVEILLANCE HOLES IN FFTF HEAT TRANSPORT CELL

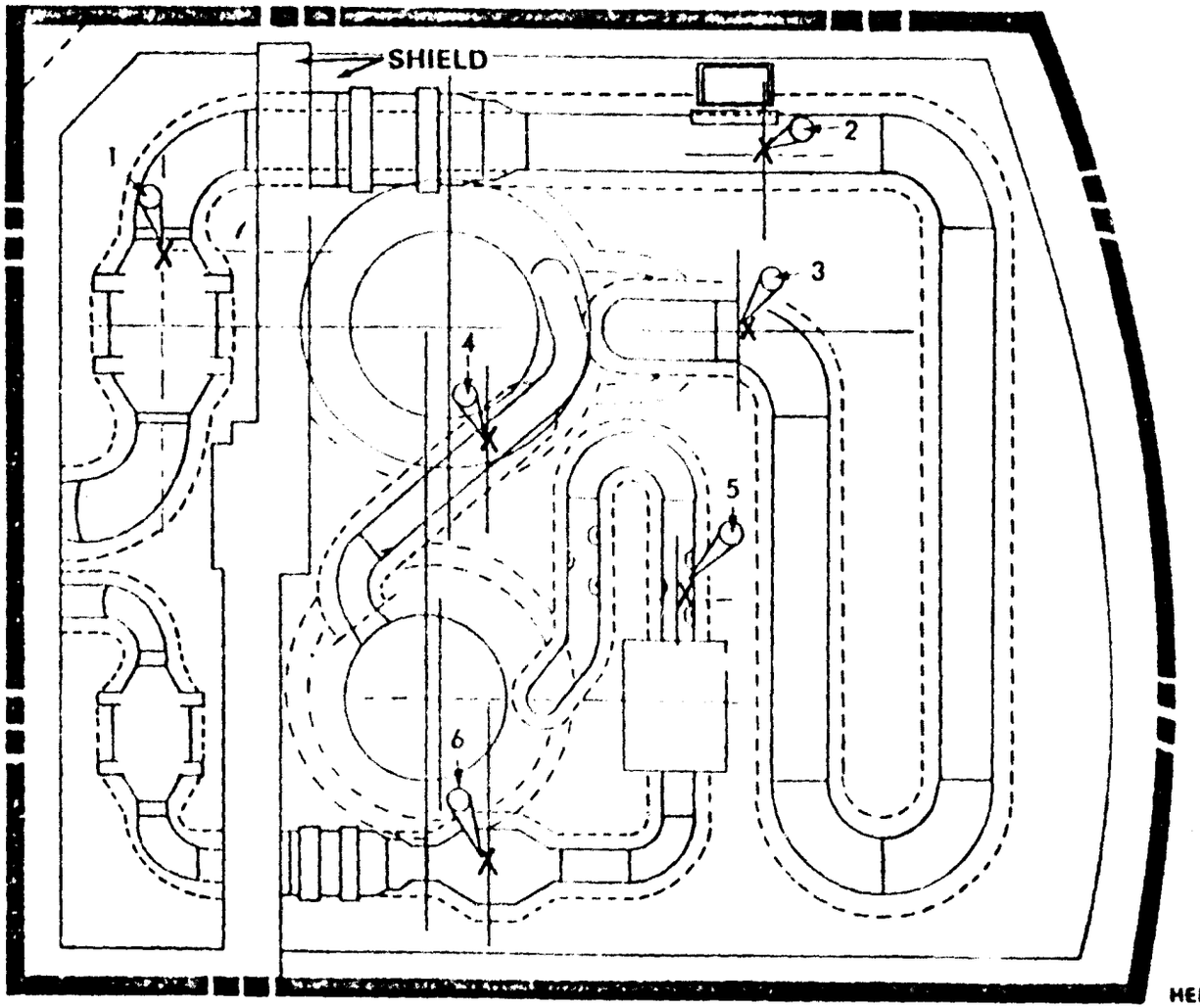


FIGURE 9