

# Technical Information on the Carbonation of the EBR-II Reactor

Summary Report Part 2: Application to  
EBR-II Primary Sodium System and  
Related Systems

Steven R. Sherman  
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March 2006



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## ABSTRACT

Residual sodium is defined as sodium metal that remains behind in pipes, vessels, and tanks after the bulk sodium metal has been melted and drained from such components. The residual sodium has the same chemical properties as bulk sodium, and differs from bulk sodium only in the thickness of the sodium deposit. Typically, sodium is considered residual when the thickness of the deposit is less than 5-6 cm. This residual sodium must be removed or deactivated when a pipe, vessel, system, or entire reactor is permanently taken out of service, in order to make the component or system safer and/or to comply with decommissioning and disposal regulations.

As an alternative to the established residual sodium deactivation techniques (steam-and-nitrogen, wet vapor nitrogen, etc.), a technique involving the use of moisture and carbon dioxide has been developed. With this technique, sodium metal is converted into sodium bicarbonate by reacting it with humid carbon dioxide. Hydrogen is emitted as a by-product.

This technique was first developed in the laboratory by exposing sodium samples to humidified carbon dioxide under controlled conditions, and then demonstrated on a larger scale by treating residual sodium within the Experimental Breeder Reactor II (EBR-II) secondary cooling system, followed by the primary cooling system, respectively. The EBR-II facility is located at the Idaho National Laboratory (INL) in southeastern Idaho, USA.

This report is Part 2 of a two-part report. This second report provides a supplement to the first report and describes the application of the humidified carbon dioxide technique (“carbonation”) to the EBR-II primary tank, primary cover gas systems, and the intermediate heat exchanger. Future treatment plans are also provided.



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# **Technical Information on the Carbonation of the EBR-II Reactor**

## **Summary Report Part 2: Application To EBR-II Primary Sodium System and Related Systems**

### **1. INTRODUCTION**

The Experimental Breeder Reactor-II (EBR-II) was an unmoderated, heterogeneous, sodium-cooled fast breeder reactor operated by Argonne National Laboratory – West, now part of the Idaho National Laboratory, in southeastern Idaho. EBR-II was a pool reactor, with the reactor core, primary sodium pumps, and the intermediate heat exchanger (IHX) submerged in a tank of molten sodium metal. When it operated, the nominal power output of the reactor was 62.5 MW thermal and approximately 20 MW electrical. The reactor began operation in 1964 and operated until final reactor shutdown in 1994. During its lifetime, the reactor served as a test facility for fuels development, hardware development, materials irradiation, and system and control theory testing.

In a pool reactor, sodium metal from the pool is pumped through the core of the nuclear reactor and out again to the pool to remove the heat of nuclear reaction. A heat exchanger internal to the pool (the intermediate heat exchanger or IHX) was used to transfer thermal energy to the secondary sodium cooling system before the sodium metal was re-injected into the pool. A schematic of the sodium pool and reactor components for EBR-II is shown in Figure 1. When EBR-II was operational, the sodium pool contained approximately 340,000 liters (90,000 gallons) of sodium metal.

In October 1994, the U.S. Department of Energy terminated the U.S. Integral Fast Reactor (IFR) Program, which included EBR-II. At that time, Argonne—West was directed to place EBR-II into a radiologically and industrially safe condition until full decontamination and decommissioning activities could be initiated. Though EBR-II was already a radiologically and industrially safe facility, this new condition was defined as an intermediate state between being fully operational and a state ready for decommissioning that would allow for a reduced staffing level and reduced facility costs while still maintaining the high safety levels that had been achieved with a much larger staff and a greater complexity of safety systems when the plant was fully operational.

This task was defined and implemented by the EBR-II Plant Closure Project (PCP) and was completed in March 2002. The first phase of the project, de-fueling the reactor, was completed by December 1996. The second phase of the project, draining the bulk sodium coolant from the EBR-II secondary and primary cooling systems, was completed by the summer of 2000. The last phase of the project involved in part placing a carbonate layer (i.e., “carbonation”) on top of the residual sodium remaining in the EBR-II sodium cooling systems. The carbonate layer, it was reasoned, would help protect the residual sodium surfaces against uncontrolled air and moisture excursions, and increase the general level of safety above what could be achieved by leaving the residual sodium in an untreated state. The carbonate layer was created by treating all exposed residual sodium metal surfaces with humidified carbon dioxide, a method which, at the time, was referred to as “passivation.” This last phase of the

EBR-II Plant Closure Project was completed in March 2002. After project completion, the EBR-II reactor was placed under a static blanket of dry carbon dioxide.

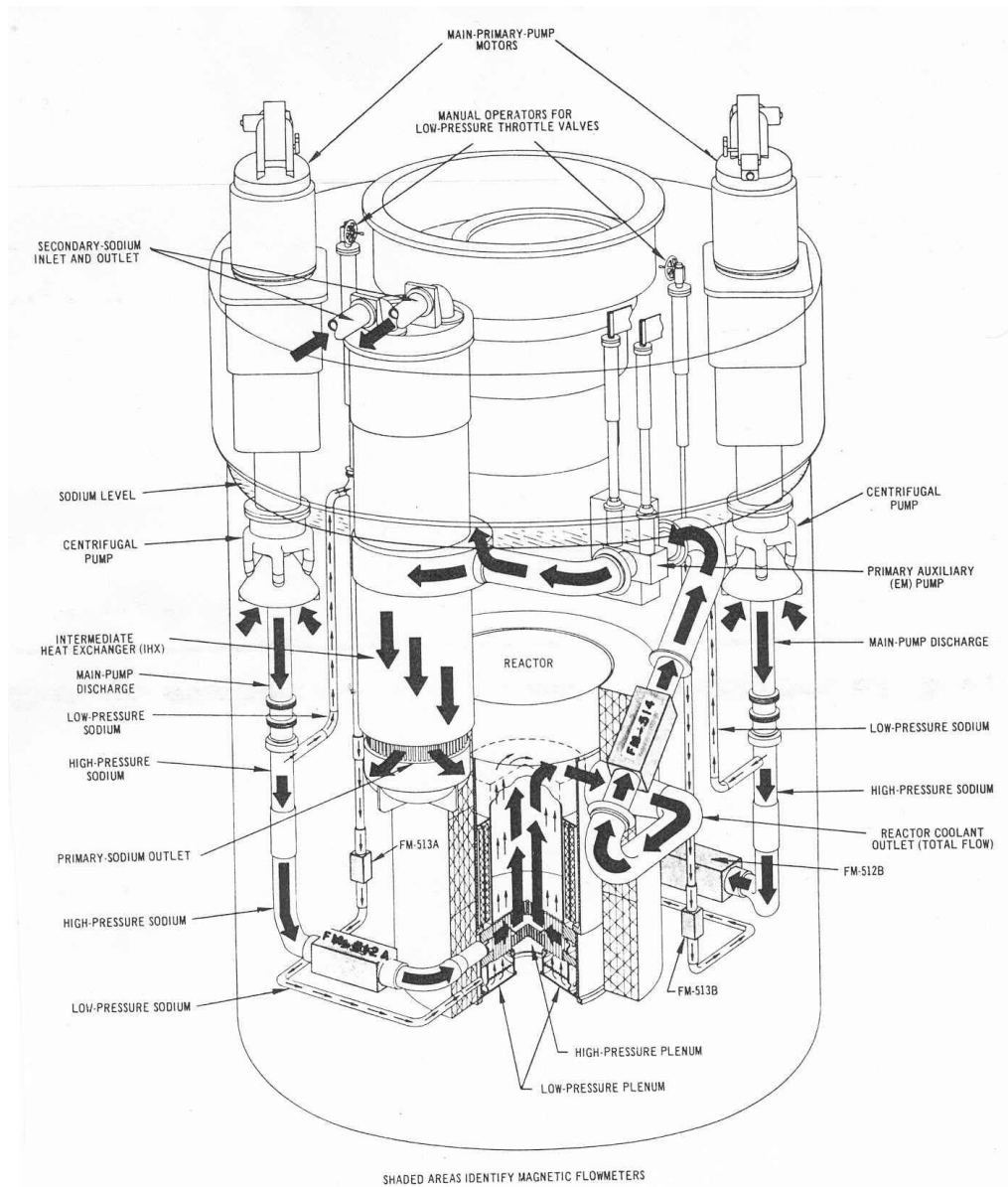


Figure 1: Schematic of EBR-II Primary Sodium Cooling System

Much more was involved in the third phase of the EBR-II Plant Closure Project than placing a carbonate layer on top of the residual sodium. Additional activities included the removal or dismantling of unessential electrical and mechanical systems, the de-energization of electrical panels, circuit breakers and instruments, and the re-working of engineering drawings to reflect these changes. Detailed historical documents called “lay-up plans” were created for all of the EBR-II sub-systems that describe the sub-system, current state of each sub-system, and the amounts of residual sodium, radioactive materials and other regulated substances remaining in the sub-systems. Since it was unknown how long each sub-system would have to remain in a static state before further decontamination or decommissioning



activities could be initiated, the “lay-up” plans were seen as a method for communicating the details and the hazards of each sub-system forward in time without having to rely on other project documents or sometimes unreliable personal recollections to communicate such information.

Though the details of the EBR-II Plant Closure Project are interesting and relevant to determining how any sodium-cooled reactor might be prepared for decommissioning, this report will not describe all of those details and will instead focus on specific actions and results associated with treatment of the residual sodium within the EBR-II primary sodium cooling system and related systems.

This report is Part 2 of a two-part report (Ref. 1). While the Part 1 report described the laboratory development of the humidified carbon dioxide treatment technique (Ref. 2) and its application to the EBR-II secondary sodium cooling system, this report describes the experience of applying the treatment technique to the EBR-II primary tank and related sub-systems. In this report, the steps taken to prepare the EBR-II primary tank for carbonation are described. Then, results and analyses are provided for the treatment of the primary tank and argon cover gas system components. The treatment results for the intermediate heat exchanger are also provided. Lastly, the future treatment path of the EBR-II primary tank and other systems is described.

## 2. PREPARATION FOR TREATMENT

Preparation for treatment of the EBR-II primary sodium cooling system included cataloging the residual sodium locations and residual sodium volumes in the EBR-II primary system, the installation of additional pressure control and instrumentation equipment on the EBR-II primary tank, the installation of a large carbon dioxide tank, allowing the primary tank to cool to near ambient conditions, and purging the primary tank system with dry carbon dioxide to remove the argon cover gas. The motivation for each step and the details of their execution are described below.

### 2.1 Catalog of Residual Sodium Locations

As a first step in treating the residual sodium within the EBR-II primary sodium system, it was necessary to determine approximately how much residual sodium remained in the system after draining the bulk sodium metal. It was known that the EBR-II primary system contained approximately 340,000 liters of sodium metal when operating, but the precise number was not known, especially after 30 years of operation in which sodium was lost from the system through evaporation and physical losses from the removal of sodium-coated parts (e.g., fuel assemblies). It was also known that approximately 338,000 liters of bulk sodium metal were drained from the EBR-II primary tank during the EBR-II Plant Closure Project, but the precision of this measured volume was not very good and was known only to within 1000 liters. By taking the difference between these two large numbers, the amount of residual sodium remaining in the EBR-II primary tank was determined to lie between 1100 and 4200 liters.

In an effort to determine more precisely the amount of residual sodium remaining in EBR-II, it was decided to rely on a geometric calculation instead. The internal geometry of the EBR-II primary tank is known precisely from engineering drawings, and the capacity of the internal structures to retain liquid sodium metal after the bulk sodium metal has been drained could be determined. The engineering drawings for all of the structures within the EBR-II primary tank were studied, hydraulic low points were identified, and the potential amount residual sodium that could have been stored at each hydraulic low point was quantified. The amount of sodium at each location was then summed to calculate the total amount of residual sodium stored in the primary tank.

For instance, it was known that a suction tube was used to draw out the bulk sodium, and that the suction tube descended to a height of 0.95 cm above the lowest point in the primary tank (the primary tank floor). Therefore, the level of residual sodium remaining on the bottom of the EBR-II primary tank must be no higher than 0.95 cm. The diameter of the tank bottom was known from the engineering drawings, and the surface area of the tank bottom could be calculated. Multiplying the tank bottom surface area by the depth of residual sodium produced a volume of sodium on the tank bottom. By this calculation, there was approximately 473 liters (125 gallons) of residual sodium on the tank bottom.

Similar calculations were repeated at other locations in the primary tank, except that the height of the residual sodium at other locations was determined by observing the heights of the drainage paths. It was assumed that sodium drained by gravity to the bottom of the primary tank as the bulk sodium was withdrawn except in places where there were no drainage outlets (i.e., local minima).

Vertical surfaces were assumed to have negligible sodium metal retention based upon visual observation of the inside of the EBR-II primary tank with video cameras. Video imaging of the primary tank walls showed clean metal surfaces with no visible sodium metal deposits.

Though it could not be strictly quantified, a certain amount of residual sodium was assumed to be on the bottom of the EBR-II primary tank cover due to the condensation of sodium metal aerosol during

regular operation of the nuclear reactor. This amount of residual sodium was assumed to be approximately 189 liters (50 gallons).

For a complete accounting of the residual sodium locations in the EBR-II primary tank prior to beginning the treatment operation, see Table 1. The total amount of residual sodium in the EBR-II primary tank was estimated to be 1116 liters (approximately 300 gallons) prior to treatment, and is consistent with the lower end of the range of values calculated from the bulk sodium measurements.

Also shown in the table are comments made about whether there are restrictions to the amount of residual sodium that could be reacted by carbonation at each location depending upon the depth of the sodium deposit, and presence of any gas-space limitations. Treatment of sodium metal beyond 5 cm depth in the laboratory was shown to be exceedingly slow due to mass transfer limitations. Therefore, little progress was expected beyond 5 cm at any particular residual sodium location in the EBR-II primary tank. Also, as was noted in the laboratory experiments, the sodium bicarbonate layer expands as it grows, and assumes an approximate 5-times larger volume than the sodium metal deposit from which it was created. If the gas space above the sodium deposit is limited, the layer will grow until the gas space is fully occupied by sodium bicarbonate, and further treatment of the residual sodium in that location will be severely curtailed.

In the table, it can be seen that there will be residual sodium remaining in the EBR-II primary tank after treating the system with humidified carbon dioxide. The humidified carbon dioxide technique will not be capable of treating 100% of the residual sodium due to geometric restrictions. Other treatment methods will need to be employed to further react sodium at these locations.

Also shown in the table are heights of the hydraulic drainage paths of the residual sodium deposits. If it is decided to fill the EBR-II primary tank with liquid water to react the remaining sodium deposits and dissolve the sodium bicarbonate reaction products, the heights of the hydraulic drainage paths will be important to know when planning the safe execution of the water fill process. With differing heights of the residual sodium deposits, only select locations are available to react with liquid water for any given height of the fill water. By carefully controlling the fill rate and fill volume as a function of residual sodium deposit height, the amount of sodium available to react with liquid water can be controlled.

According to the table, there were 1116 liters (300 gallons) of residual sodium prior to beginning carbonation and the maximum amount of sodium that could be reacted using carbonation is 884 liters (236 gallons). It will not be possible to react all of the residual sodium within the EBR-II primary tank with carbonation, and a minimum of 232 liters (64 gallons) of residual sodium will remain in the tank at various locations after carbonation is stopped.

Table 1. Residual Sodium Deposits in EBR-II Primary Tank

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
1	Low pressure plenum	27	Annular pool, 3.2 cm deep in 35.6 cm tall space. 207.6 cm O.D., 176.5 cm I.D.	No	Limited gas circulation.	0	N/A	EB-1-26800 EB-1-25812
2	High pressure plenum	125	Annular pool, 8.7 cm deep in 27.9 cm tall space. 218.1 cm O.D., 67.6 cm I.D.	Yes	Flow closed off into plenum prior to full treatment. Limited gas circulation.	66	124.5	EB-1-26800 EB-1-25812
3	Inlet pipes to high pressure plenum	117	Pool, 9.4 cm deep in bottom of 30.5 cm diameter pipe and fittings	Yes	Flow closed off into high pressure plenum prior to full treatment, so gas flow becomes static once plenum limit is reached.	49	124.5	EB-1-25231
4	High pressure plenum inside flow distributing ring and outside Row 7 step in lower grid plate	42	Pool, 4.9 cm deep in 27.9 cm tall space. 167.8 cm O.D., 66.0 cm hex., penetrated by 510 tubes.	No	Limited gas circulation.	0	N/A	EB-1-26800

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
5	Between blanket lower adapter and sleeve between grid plates	0	510 annular volumes, 27.9 cm deep, between sleeves (4.0 cm I.D.) and lower adapters (3.7 cm O.D.). Very tight annulus around adapters (0.02 cm width).	Yes	Limited gas circulation.	0	N/A	EB-1-26800
6	Control rod position dummy assembly	0	Max. depth of 162.6 cm, located above grid plate between S/A hex and guide tube hex (0.38 cm gap).	Yes	Subassemblies were removed. Limited gas circulation.	0	N/A	
7	Inner shield region between inner and outer walls and outlet pipe	11	4.6 cm deep in 142.2 cm tall space. (Height to outer drain hole in shield-ring can support ring)	No	Limited gas circulation.	0	N/A	EB-1-26800

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
8	Inner shield region between thermal baffle and outer wall	11	Annular pool, 132.0 cm max. depth, 228.0 cm O.D., 225.4 cm I.D. Unknown surface area and depth.	Yes	Assume 10% of volume remains after original draining. Small exposed surface area to gas space will limit sodium reaction.	11	165.1	EB-1-26800
9	Top flange of reactor vessel	15	Cavity around top of reactor vessel. Approx. 3.8 cm x 5.7 cm x 221 cm diameter. Open at top through 0.48 cm wide by 3.18 cm deep slot.	Yes	No room for expansion of carbonate reaction product. Growth of layer will plug access slot.	15	381.0	EB-1-26111
10	Reactor cover thermal baffles	11	Two horizontal plates, 209.6 cm O.D. spaced 0.71 cm apart.	Yes	Assume 40% of original sodium remains after draining. Very little exposed surface area.	11	652.8	EB-1-26799 EB-1-26800

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
11	Sleeves and bellows for gripper, auxiliary gripper and hold down	11	Top and bottom bellows and annular gap (0.36 cm) between sleeves and cover penetrations	Yes	Restricted space and access. Limited gas circulation	11	657.9 to 739.1	EB-1-28815
12	Sleeves and bellows for control rod drives	8	Bellows for control rod penetrations assembly	Yes	Restricted access. Limited gas circulation.	8	657.9 to 739.1	EB-1-27113 EB-1-27156
13	Guide funnels for control rod drives	38	Complex geometry inside flow baffle and outside guide funnels of control rod drive penetration assembly	Yes	Restricted access. Limited gas circulation.	38	739.1	EB-1-27127

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
14	Outside flow baffle around gripper/hold down	11	Pool, 13.3 cm deep, outside flow baffle and inside recessed area around gripper and holddown. Open to tank.	Yes		6	739.1	EB-1-27141 EB-1-27142
15	Inside flow baffle around gripper/holddown	None	Pool, 21.6 cm deep, inside flow baffle and outside gripper holddown shafts. Open to tank.	No	This is assumed to have drained with the bulk sodium.	None		EB-1-27699 EB-1-27101
16	Recessed area around lifting columns	8	2 pools, 6.4 cm deep, inside recessed area of lifting-column attachment pads.	No		0		EB-1-27141 EB-1-27145
17	Safety rod drive lift tubes	<1	2 cylindrical pools, 2.28 cm diam. X 55.9 cm deep. Restricted access.	Yes	Limited circulation.	<1	83.8	



Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
18	Transfer arm pedestal	4	10.1 cm X 457 cm pipe in 20.3 cm socket. Sodium should drain down to socket. Located on top of pedestal is a 15.2 cm diam. X 14.6 cm long pipe with no drain.	Yes	Limited circulation.	<4	76.2 to 464.8	EB-1-28269
19	Pressure transmitting piping	8	1.3 cm pipe, sch. 40 and sch. 80, 34.5 m length.	Yes	No drainage expected.	8		
20	Heater guide funnels	2	4.8 cm i.d. pipe. Depths of 15.9 cm, 36.2 cm, 70.5 cm.	Yes	No drainage expected.	2	73.7, 94.0, 127.0	ID-2D-11344
21	Auxiliary pump bellows	2	Cylindrical sector, 30.5 cm o.d., 18.1 cm i.d., 6.4 cm long.	No	Limited circulation.	0		EB-1-25274 EB-1-25438

Location	Location Description	Deposit Volume (liters)	Location Geometry	Treatment Limitations? (Yes/No)	Treatment Comments	Unreacted Sodium Expected After Treatment (liters)	Vertical Height of Deposit above Tank Bottom (cm)	Drawing Reference Number
22	Pipe supports	None	Framework constructed from 10.2 cm, 5.1 cm, and 2.5 cm pipe which supports reactor inlet piping. Location of drainage holes not defined. Restricted access.	No	Pipe supports are believed to have drainage holes and probably drained. These may contain some internal sodium deposits of unknown quantity.	None		EB-1-27179
23	Primary tank bottom	473	0.95 cm depth x 792 cm diam. Open to tank.	No		0		
24	Primary tank cover	189	Open to tank. Aerosols in annulus areas. Sodium in rotating plug.	Yes	Limited circulation.	0		
Total estimated sodium (liters)		1116						
Maximum sodium amount available to be carbonated (liters)		884						
Minimum amount remaining after carbonation (liters)		232						

Apart from the EBR-II primary tank, there are four components that were treated separately. These components are the intermediate heat exchanger, the Vapor Traps #1 and #2 (part of the EBR-II argon pressure control system), and the Controlled Temperature Profile (CTP) condenser (also part of the EBR-II argon pressure control system). The amounts of residual sodium in these components is not well characterized, though it is believed that approximately 45 gallons (200 liters) of residual sodium may remain in the bottom of the intermediate heat exchanger, and tens of kilograms may reside inside the vapor traps and the CTP condenser in the form of sodium aerosol.

## 2.2 Pressure Control Equipment

The EBR-II primary tank cover has approximately 58 nozzles or penetrations (Figure 2) through which instruments and devices were inserted to operate and monitor the reactor. One of these nozzles contained a device for removing failed fuel elements from the reactor core. The failed fuel removal system was never used, and it was decided to remove the system from the nozzle and to use the nozzle as a gas outlet port during carbonation. An exhaust gas pipe was attached to the nozzle, and the pipe was connected to the general HEPA-filtered suspect exhaust stack for the EBR-II facility. This change converted the tank from a “closed” system to an “open” system.

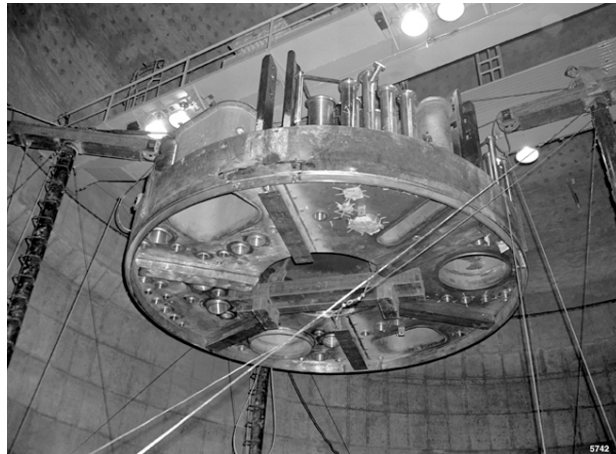


Figure 2. Bottom of EBR-II Tank Cover Showing Large and Small Nozzle Penetrations

A mechanical back-pressure regulator was installed in the gas outlet line from the primary tank. The back-pressure regulator was set to open between 250 Pa and 1000 Pa (1” and 4” H<sub>2</sub>O) so that the pressure inside the primary tank had to exceed the valve threshold pressure before it would open. Otherwise the valve stayed in the closed position. The back-pressure regulator was installed in order to prevent the flow of air from the exhaust stack into the EBR-II primary tank in the event the pressure inside the tank fell below the pressure in the exhaust stack.

The primary pressure protections systems for the EBR-II primary tank were also maintained during carbonation. These systems include an oil-filled floating head tank to adjust for minor pressure changes (adjustments on order of 100’s of Pa), a back-up manual pressure control system (adjustments on order of 10’s of kPa), and a primary tank rupture disk (large overpressure events), which was part of the manual pressure control system. The activation threshold for changes in the floating head tank had to be adjusted so that the system did not conflict with the primary tank back-pressure regulator. The back-pressure regulator was given precedence, so that the preferred pathway for treatment gases exiting the

EBR-II primary tank was through the back-pressure regulator and to the exhaust stack, rather than through the floating head tank system.

No adjustments were made to the manual pressure system and the rupture disk for the primary tank, and they were maintained at the same action levels as during reactor operation.

## **2.3 Instrumentation**

Instrumentation was installed on the tank side of the back-pressure regulator to monitor the composition and flow rate of the exit gases from the primary tank. A Teledyne Analytical Instruments #235B thermal conductivity detector was installed to measure hydrogen concentration in the off-gas and was calibrated for the range 0 to 5 vol%. A Teledyne Analytical Instruments #326RB oxygen monitor was installed to measure for oxygen in-leakage and was calibrated to read in the range from 0 to 1 vol%. A Panametrics #MCHTR-1 moisture monitor with remote moisture probe was installed to measure the relative humidity of the exhaust gas. Also, a Fluid Components International GF92 thermal conductivity mass flow meter was installed to measure the mass flow of exhaust gas.

A safety action level of 0.5 vol% oxygen was established. If the measured oxygen level exceeded 0.5 vol% while treatment of the EBR-II primary tank was being performed, the flow of humidified carbon dioxide to the primary tank system would be stopped and replaced with dry carbon dioxide until the measured oxygen concentration fell below 0.5 vol%.

A safety action level of 4.0 vol% hydrogen was also established. If measured hydrogen concentration rose above 4.0 vol% during treatment, the flow of humidified carbon dioxide was stopped and replaced with dry carbon dioxide until the condition subsided.

Both action levels were established to avoid the creation of potentially hazardous atmospheric conditions inside the EBR-II primary tank. While the emergence of such conditions in the primary tank do not mean that a hydrogen conflagration or explosion is imminent, such conditions are necessary for the occurrence of such events. Switching the gas flow from humidified carbon dioxide to dry carbon dioxide above the action thresholds stops the further flow of moisture into the tank while still maintaining gas flow through the system. Gas flow is maintained to purge hydrogen from the system in order to eliminate any hydrogen combustion hazard.

## **2.4 Installation of Carbon Dioxide Tank**

A liquid carbon dioxide tank was installed adjacent to the EBR-II facility to supply carbon dioxide on a continuous basis to the facility during residual sodium treatment. The tank has a 14,000 lb (6400 kg) carbon dioxide capacity, and was installed on a permanent cement pad. The tank was sized to supply carbon dioxide at a rate of 5 standard cubic feet per minute (1 scfm = 1 ft<sup>3</sup> per minute at 60 °F and 1 atm) without requiring a refill for at least 2 weeks.

## **2.5 EBR-II Primary Tank Cooling**

When the bulk sodium was drained from the EBR-II primary tank, the primary tank was maintained at approximately 200 °C (392 °F) to keep the bulk sodium in a molten state. Bayonet heaters internal to the primary tank were used to supply heat to the bulk sodium until the sodium level in the primary tank fell below the height of the bottom edge of the bayonet heaters, and then heat was supplied from hot air blowers that forced heat air passed the primary tank outer wall. With the combination of

internal and external heaters, technicians were able to maintain the temperature set point until all of the bulk sodium had been drained.

After drainage was complete, the internal heaters were deactivated, and the heat elements in the external blowers were deactivated, and the primary tank was allowed to cool by radiant heat losses, and by natural and forced convection of the surrounding atmosphere. The primary tank cooled slowly, and it took several months for the primary tank temperature to fall to below 50 °C (122 °F), the temperature at which the earlier sodium/carbon dioxide experiments were conducted in the laboratory. While the primary tank was cooling, argon was maintained as the cover gas. Since the blowers still provided some thermal energy to the primary tank from friction losses, the blowers were maintained in the active position throughout carbonation in order to keep the temperature of the primary tank above room temperature. This was done to avoid condensation of moisture inside the primary tank during carbonation. As a result, the temperature of the primary tank did not fall below 40 °C (104 °F) during treatment.

## **2.6 Cover-gas Changeover**

Once the EBR-II primary tank had cooled to near ambient conditions, the cover gas in the primary tank was converted from argon to carbon dioxide. Though such a changeover could be done concurrently with the influx of humidified carbon dioxide and the outflow of exhaust gases from the primary tank during treatment, the EBR-II primary tank is large enough that it would take days to purge a majority of the argon from the tank at the input rate of 5 scfm. During such a changeover process, the moisture in the humidified carbon dioxide would still be available to react with residual sodium, and sodium hydroxide could accumulate instead of reacting immediately to form sodium bicarbonate. While sodium hydroxide is not directly hazardous, it can cause operational and safety problems because it is hydrophilic. It will absorb moisture from the treatment gas and reduce its effectiveness, and, if enough moisture is absorbed, it can become a liquid solution (50 wt% NaOH). Such events occur when residual sodium is treated with humidified nitrogen, a process that can be affected by occasional uncontrolled sodium-water reactions. During the initial stages of introducing humidified carbon dioxide into a primary tank filled with argon, the atmosphere in the primary tank is mostly argon, and the treatment process will behave like a humidified inert gas treatment process until the cover gas changeover has been largely performed.

To avoid such problems, the primary tank was purged with dry carbon dioxide for approximately 11 days before introducing any moisture to the system. According to calculations, purging the system with dry carbon dioxide for this time period would remove greater than 99% of the argon from the system (assuming the system was well mixed).

No measurements were performed to verify the effectiveness of the purge.

## **2.7 Carbon Dioxide Humidification Cart**

The humidification cart used to treat the EBR-II secondary sodium system was used again to treat the EBR-II primary sodium system. The cart (it is so called because the equipment is placed on a pushcart) is designed for a supply flow rate of 134 standard liters/minute (5 scfm) of carbon dioxide. To provide this flow rate, the humidification cart requires a regulated carbon dioxide supply (approximately +138 kPa-gauge) from a dedicated source. A 170-liter clear acrylic water tank is used to contain the liquid water that is used to humidify the carbon dioxide. In the bottom of the tank are four stainless steel sintered metal sparging elements. The carbon dioxide gas stream flows through these sintered elements and bubbles through the water tank. A portion of the humidified gas flows through a Panametrics

#MCHTR-1 with remote moisture probe monitor to measure the relative humidity of the outgoing gas stream. The relative humidity is displayed on a digital indicator on the cart control panel. A by-pass line around the water tank allows the operator to control the relative humidity of the gas stream by controlling the relative flow split between dry and humidified carbon dioxide. The cart also contains other valves, mechanical flow meters and pressure indicators. Photographs of the cart are shown in Figure 2.

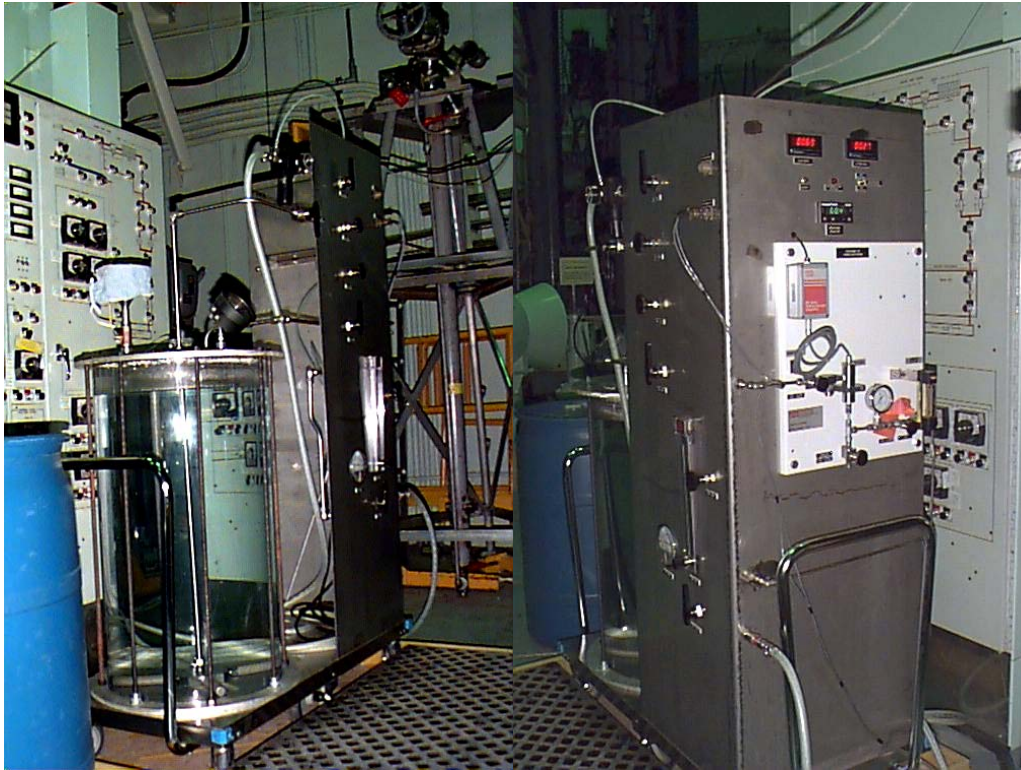


Figure 3. Carbon dioxide humidification cart, from two vantage points.

### 3. CARBONATION OF EBR-II PRIMARY SODIUM SYSTEM

#### 3.1 EBR-II Primary Tank

##### 3.1.1 Initial Carbonation: Plant Closure Project Experience

The initial experience of carbonating the residual sodium within the EBR-II primary tank occurred during the EBR-II Plant Closure Project. Carbonation occurred between 9:00 11 December 2001 and continued with one interruption until 8:50 4 February 2002.

Humidified carbon dioxide flowing at 134 slm (5 scfm) was introduced to the primary tank through the sodium suction tube used to withdraw bulk sodium from the tank. By using this tube, humidified carbon dioxide was introduced into the bottom of the primary tank, while exhaust gases were vented through the top of the primary tank. A graph of the measured hydrogen concentration and relative humidity of the exhaust gas during this treatment period is shown in Figure 4.

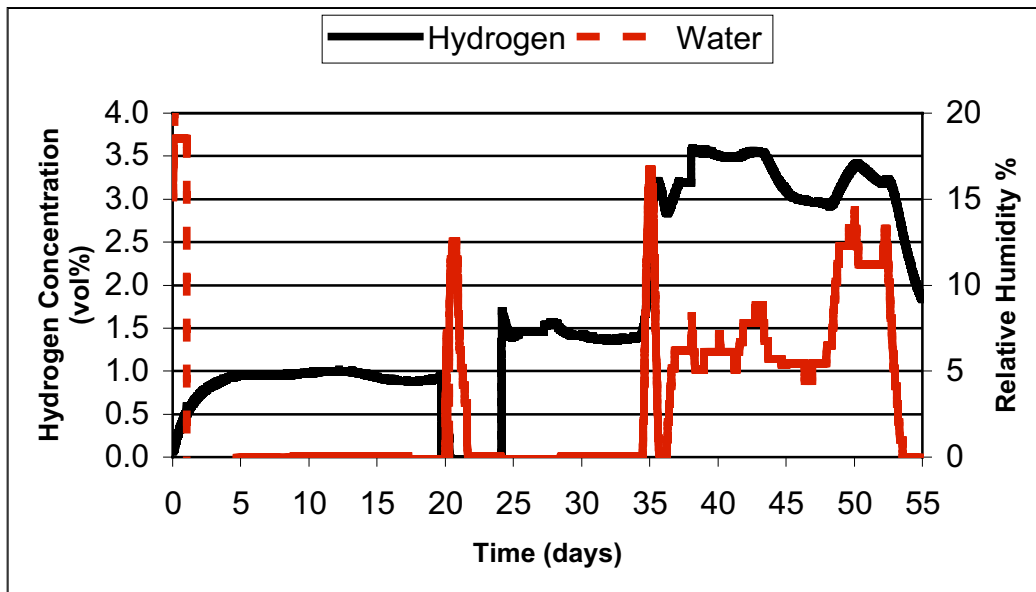


Figure 4. Measured H<sub>2</sub> Concentration and Relative Humidity in Exhaust Gas

A comparison of the measured hydrogen concentration with the exhaust gas mass flow rate is shown in Figure 5.

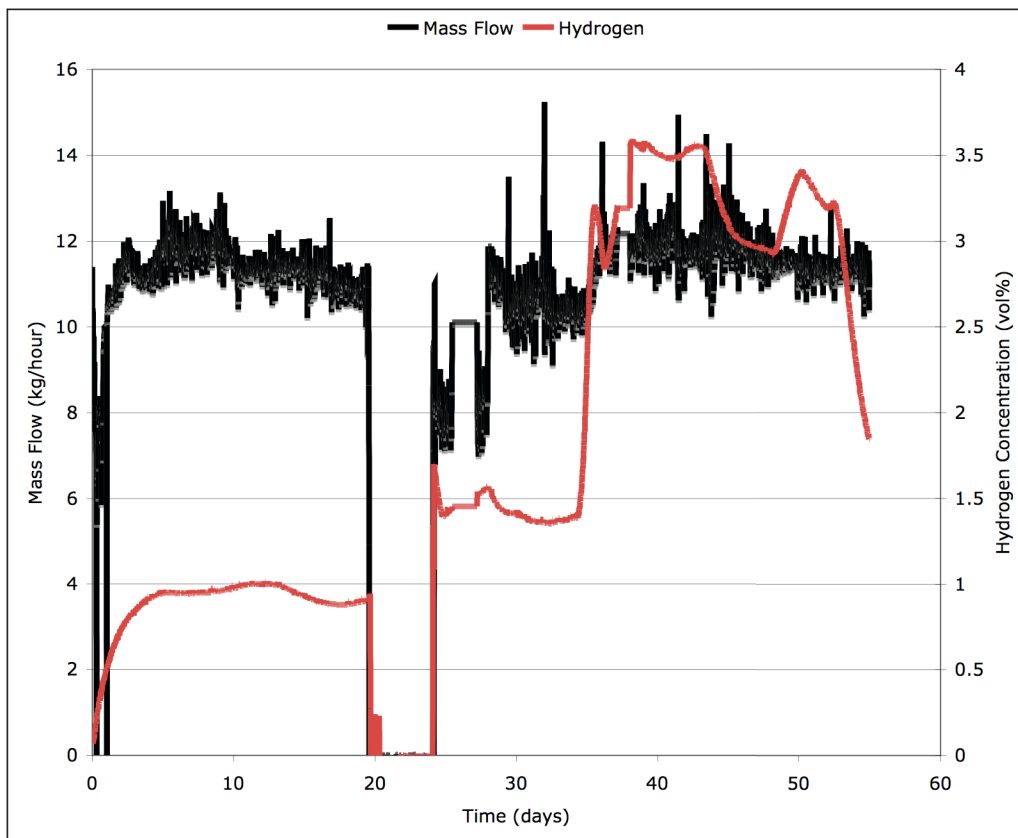


Figure 5. Measured H<sub>2</sub> Concentration and Mass Flow Rate in Exhaust Gas

The figure is divided into three regions. The first region on the left shows the measured hydrogen concentration when carbonation was initiated (time span 20 days). The water temperature in the humidification cart was measured at 18 °C and the relative humidity of carbon dioxide being generated by the cart was approximately 70%. The measured hydrogen concentration of 1 vol% is consistent with the concentration of hydrogen that would be produced if 100% of incoming moisture were consumed by residual sodium within the primary tank.

After the first 20 days, there was an interruption in treatment for 4 days. At this time, the carbon dioxide tank ran out of carbon dioxide, and flow of carbon dioxide to the humidification cart and the primary tank back-up pressure control systems stopped. This was not a controlled shutdown, and no active pressure control system was in place to keep the differential pressure in the primary tank from “floating” with changes in atmospheric pressure. Fortunately, the back-pressure regulator operated properly and sealed the primary tank exhaust gas line. No hydrogen was measured during this time period, which supported the no-flow condition in the exhaust line. The spike in relative humidity during this time is unexplained, since it appears to be of shorter duration than the interruption, and was not accompanied by a spike in the measured oxygen concentration as would be encountered if air were leaking past the back-pressure regulator.

The carbon dioxide tank was refilled and the flow of carbon dioxide to the primary tank was restored 4 days later. Over the next ten days (region 2), the measured hydrogen concentration was approximately 1.5 vol%. No changes had been made to the humidification cart settings (i.e., temperature, flow rate), so the moisture input hadn’t changed from prior to the interruption. Since a hydrogen



concentration of 1 vol% was expected, either additional moisture was being introduced into the primary tank from another source (not probable), or the hydrogen meter was incorrect (probable). A re-calibration of the hydrogen meter was required to check the accuracy of the hydrogen meter, but this could not be done without interrupting the carbonation operation. It was decided to continue carbonation without recalibrating the hydrogen meter.

The third region spans from day 35 through day 55. In this region, the water tank on the humidification cart was heated from 18 °C to approximately 32 °C in order to increase the moisture content of the carbon dioxide and increase the treatment rate. This increase in temperature resulted in increased hydrogen concentration in the exhaust gas (3.0-3.5 vol%) and an increase in exhaust gas relative humidity. Measurable humidity in the exhaust gas at the higher moisture input rates suggested that the reaction of sodium metal within the primary tank had become surface diffusion limited, which may have been indicative of the formation of surface layers of sodium bicarbonate on the available sodium surfaces.

A mass balance was the tool used to assess the effectiveness of the treatment process, and was performed in two ways. The first way was to integrate the hydrogen and water concentration data and mass flow data from the exhaust gas instruments and relate this information to the amount of sodium metal reacted. The second method was to measure how much water had been evaporated from the humidification cart, and make the simple assumption that all of the water evaporated from the humidification cart reacted with sodium metal inside the primary tank. An integration of the exhaust gas data is more precise because it takes into account variations in humidity, oxygen and hydrogen concentration, and mass flow changes, but the water evaporation data is very simple to collect and provides an upper limit on the amount of sodium that could have possibly reacted.

In Region 1 of the figures above, an integration of the measured exhaust gas data showed that 49 kg of sodium metal had been consumed. According to the laboratory records, 36 liters of water were evaporated during this time period, which would be capable of reacting 46 +/- 4 kg of sodium metal if all of this moisture reacted with sodium inside the primary tank. The two calculation methods are consistent in this region.

In Regions 2 and 3, there was reason to suspect the accuracy of the hydrogen meter, and so another comparison was made of the two methods, giving precedence to the water evaporation method for determining how much sodium metal was actually consumed in the primary tank. According to an integration of the exhaust data, an additional 232 kg of sodium had reacted, whereas the water evaporation data showed that no more than 148 +/- 13 kg of additional sodium could have reacted. These calculations support the hypothesis that the hydrogen meter calibration was not correct at this time, and that the water mass balance was needed to calculate how much sodium had reacted.

Adding up the amount of sodium reacted in Regions 1, 2, and 3 using the water evaporation method, a total of 194 +/- 17 kg (52 gallons +/- 5 gallons) were reacted during initial carbonation. This amount is  $194/1100 * 100\% = 17.6\%$  of the original residual sodium inventory in the primary tank.

### **3.1.2 Assessment of Reaction Depth and Expected Treatment Rate**

#### Reaction Depth

Reaction depth is defined as the depth of sodium reacted as measured from the original surface of residual sodium. This is important to know when assessing whether residual sodium at any particular location may have been consumed. Reaction depth cannot easily be measured directly in the primary tank due to the high radiation field in the tank and physical accessibility issues and must be calculated instead.

The reaction depth is calculated in the following manner. First, it is assumed that the gas space within the primary tank is well mixed, so that all sodium metal surfaces within the primary tank are accessible to incoming moisture. Then, it is assumed that the reaction rate of sodium at any particular location is proportional to the exposed sodium surface area at that location. This assumption has the implication that the bicarbonate layer has the same thickness on every exposed residual sodium surface, at least while the residual sodium underneath the layer is still reacting with moist carbon dioxide. Next, the information presented in Table 1 is converted into numerical data in order to determine the exposed surface area and surface fraction at each location. This numerical data is given in Table 2.

Table 2. Surface Area Parameters for Residual Sodium Deposits in EBR-II Primary Tank

Location #	Volume (cm <sup>3</sup> )	Initial Depth (cm)	Minimum Depth (cm)	Effective Exposed Surface Area (cm <sup>2</sup> )	Surface Fraction
1	27,000	3.2	0.0	8,500	0.008
2	125,000	8.7	4.9	15,000	0.014
3	117,000	9.4	3.9	12,000	0.011
4	42,000	4.9	0.0	8,500	0.008
5	0	0	0	0	0
6	0	0	0	0	0
7	11,000	4.6	0.0	2,400	0.002
8	12,000	13.2	0.0	900	0.001
9	15,000	5.7	5.7	0	0
10	11,000	0.71	0.71	0	0
11	11,000	0	0	0	0
12	8,000	0	0	0	0
13	38,000	0	0	0	0
14	11,000	13.4	7.3	800	0.001
15	0	0	0	0	0
16	8,000	6.4	0.0	1,100	0.001
17	1,000	55.9	55.9	0	0
18a	2,400	14.6	14.6	0	0
18b	1,600	20.3	20.3	0	0
19	8,000	508	508	0	0
20a	300	15.9	15.9	0	0
20b	600	36.2	36.2	0	0
20c	1,100	70.5	70.5	0	0
21	2,000	6.4	0.0	310	0.000
22	0	0	0	0	0
23	473,000	0.95	0.00	500,000	0.477
24	189,000	0.38	0.00	500,000	0.477

In the table, the minimum depth was determined by the available free space for the growth of a bicarbonate layer rather than by reaction rate limitations. In some locations, the residual sodium might be completely consumed before the bicarbonate layer height reaches its limit, and in other locations the bicarbonate layer will grow until the gas space above the residual sodium deposit is completely filled and

no further reaction of residual sodium can be performed. The effective exposed surface area was determined from the information given in Table 1 and by examining the engineering drawings. The surface fraction was determined by dividing the effective exposed surface area at any one location by the total exposed surface area. The amount of sodium reacted at any one location is calculated by multiplying the surface fraction times the total amount of sodium reacted. In this way, the mass of sodium reacted is proportioned to each location in relation to the calculated surface fractions. For example, if 1 kg of sodium is known to have reacted, then 0.54 kg (0.54 x 1 kg) reacted at location 23.

The surface fraction is re-evaluated whenever residual sodium at a particular location becomes unavailable due to the growth of the bicarbonate layer or when the residual sodium at that location is completely consumed. Additional sodium reacted is apportioned using the new surface area fraction amounts.

The relationship between sodium mass and sodium depth is shown in Table 3. Table 3 shows the mass of sodium at each location, as separated into increments of depth. The increments are not arbitrary and either correspond to the minimum depth at which the reaction rate of sodium becomes controlled by the thickness of the sodium bicarbonate layer (0.1 cm), or the maximum depths at which sodium at various locations can be reacted.

At the end of the EBR-II Plant Closure Project, approximately 194 kg of residual sodium had been reacted. This mass of residual sodium reacted corresponds to a sodium treatment depth of almost 0.2 cm and a sodium bicarbonate layer depth of almost 1.0 cm throughout the EBR-II primary tank.

Reaction Rate

Assuming that the reaction rate of residual sodium is proportional to the thickness of the bicarbonate layer above the sodium surface, a prediction of reaction rates versus sodium consumed or a projection of how long it would take to react a given mass of residual sodium within the primary tank can be calculated. From the laboratory experiments (Ref. 1), the following behavior was observed:

Layer Depth < 0.5 cm	No rate effect
Layer Depth ≥ 0.5 cm	$\text{rate (cm/day)} = \frac{0.21}{\text{NaHCO}_3 \text{ layer depth (cm)}}$

For bicarbonate layer thicknesses less than 0.5 cm, the rate at which moisture was input to the system being treated seemed to be rate controlling. Once a surface layer of sodium bicarbonate had been established (thickness greater than 0.5 cm), the bicarbonate layer thickness appeared to become rate controlling. The information given in Tables 2 and 3 is used to convert between mass of sodium reacted and the depth of sodium reacted at each location, which is directly proportional to the height of the bicarbonate layer above the sodium and inversely proportional to the reaction rate.

Table 3. Mass of Residual Sodium in Primary Tank by Location and Sodium Depth

Location	Total Mass (kg)	Available Mass (kg)	Sodium Mass Reacted (kg) Per Sodium Depth Increment (cm)												Total Reacted (kg)	Remain (kg)		
			0.00 to 0.100	0.100 to 0.381	0.381 to 0.953	0.953 to 3.18	3.18 to 3.65	3.65 to 3.81	3.81 to 4.29	4.29 to 4.60	0.00 to 0.100	0.100 to 0.381	0.381 to 0.953	0.953 to 3.18			3.18 to 3.65	3.65 to 3.81
1	26	26	0.82	2.3	4.7	18.2											26	0
2	123	51.2	1.4	3.9	8.0	31.2	6.7										51.2	71.8
3	115	52.5	1.2	3.5	7.0	27.3	5.8	1.9	5.8								52.5	62.5
4	41	41	0.83	2.3	4.7	18.4	3.9	1.3	3.9	2.6							37.9	3.1
5	0	0																0
6	0	0																0
7	11.3	11.3	0.24	0.68	1.4	5.4	1.2	0.38	1.2	0.77							11.3	0
8	12	12	0.15	0.41	0.83	3.2	0.7	0.23	0.69	0.46							6.67	5.33
9	15	0																15
10	11	0																11
11	11	0																11
12	8	0																8
13	37	0																37
14	11	3.21	0.08	0.23	0.47	1.9	0.40	0.13									3.21	7.79
15	0	0																0
16	7.41	7.41	0.12	0.33	0.67	2.6	0.56	0.19	0.56	0.37							5.40	2.01
17	1	0																1
18	4	0																4
19	8	0																8
20	2	0																2
21	1.85	1.85	0.03	0.08	0.17	0.65	0.14	0.05	0.14	0.09							1.35	0.50
22	0	0																0
23	463	464	49	137	278												464	0
24	185	185	49	137													185	0
Totals	1,095	864															845	250

Figure 6 shows long-range treatment rate and treatment time predictions assuming a humidification cart tank temperature of 25 °C and a carbon dioxide flow rate of 134 slm. The curves were generated by using the observed rate equation and the information given in Tables 2 and 3. According to the figure, treatment rates are fairly rapid at the start of the process, but begin to fall quickly as the thickness of the sodium bicarbonate layer grows inside the primary tank. The figure shows that would take almost 700 days (1.9 years) of continuous treatment using the carbonation process to react approximately 800 kg (73%) of the residual sodium within the primary tank. At this point, all of the residual sodium on the bottom of the primary tank and the bottom of the primary tank cover will have been consumed, and significant amounts of residual sodium in other less accessible locations will have been at least partially converted into sodium bicarbonate.

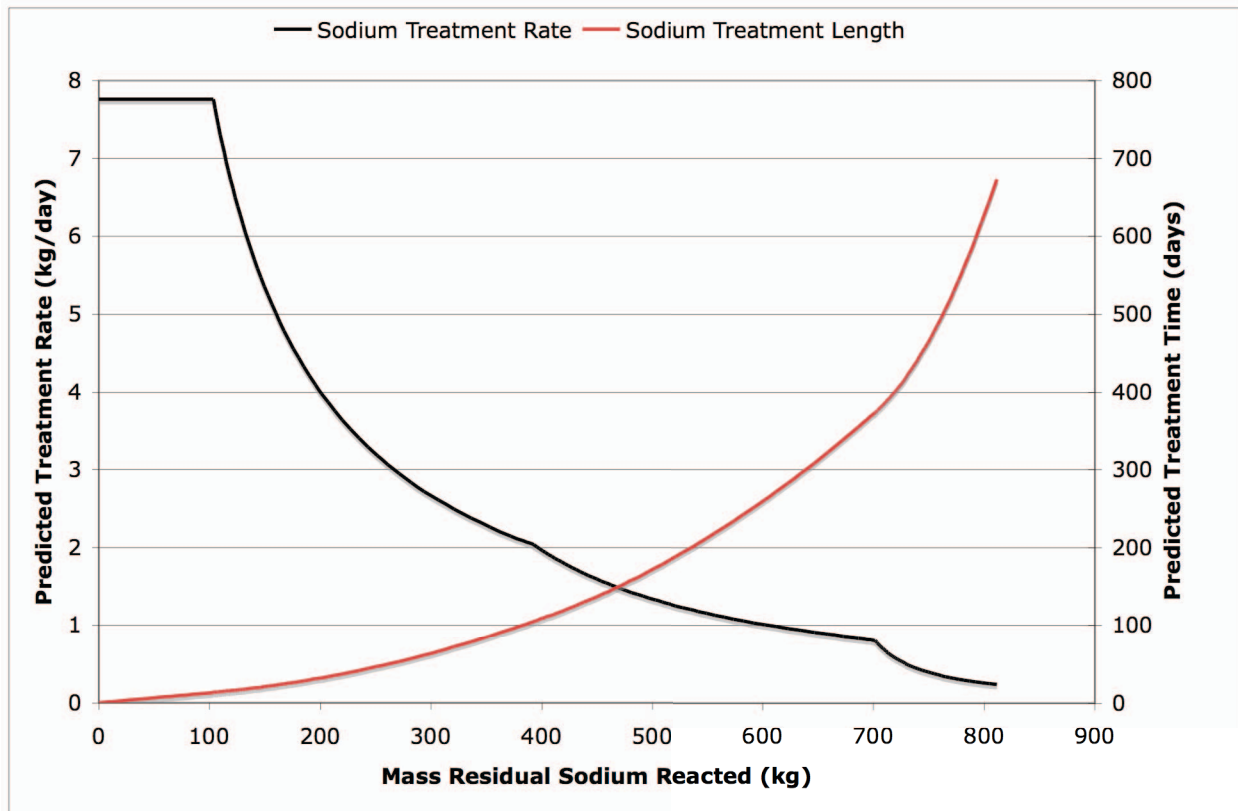


Figure 6. Predicted Treatment Rate and Total Treatment Time

Figure 7 shows the predicted treatment depth of sodium reacted and predicted sodium mass reacted versus reaction time. This figure provides another perspective on the time it takes to react the residual sodium to a certain depth or to react a certain mass of sodium within the primary tank. According to the figure, the carbonation process becomes increasingly less efficient as time progresses due to the build-up of a sodium bicarbonate layer on top of the sodium and the depletion of sodium deposits within the primary tank. The kink in the depth curve occurs when the bottom of the primary tank is depleted of residual sodium, the single largest source of residual sodium in the primary tank. Once that source is depleted, more moisture is available to react with residual sodium at other locations. The overall rate of reaction, however, continues to decline due to the increasing thickness of the sodium bicarbonate layer and the decreasing overall residual sodium surface area.

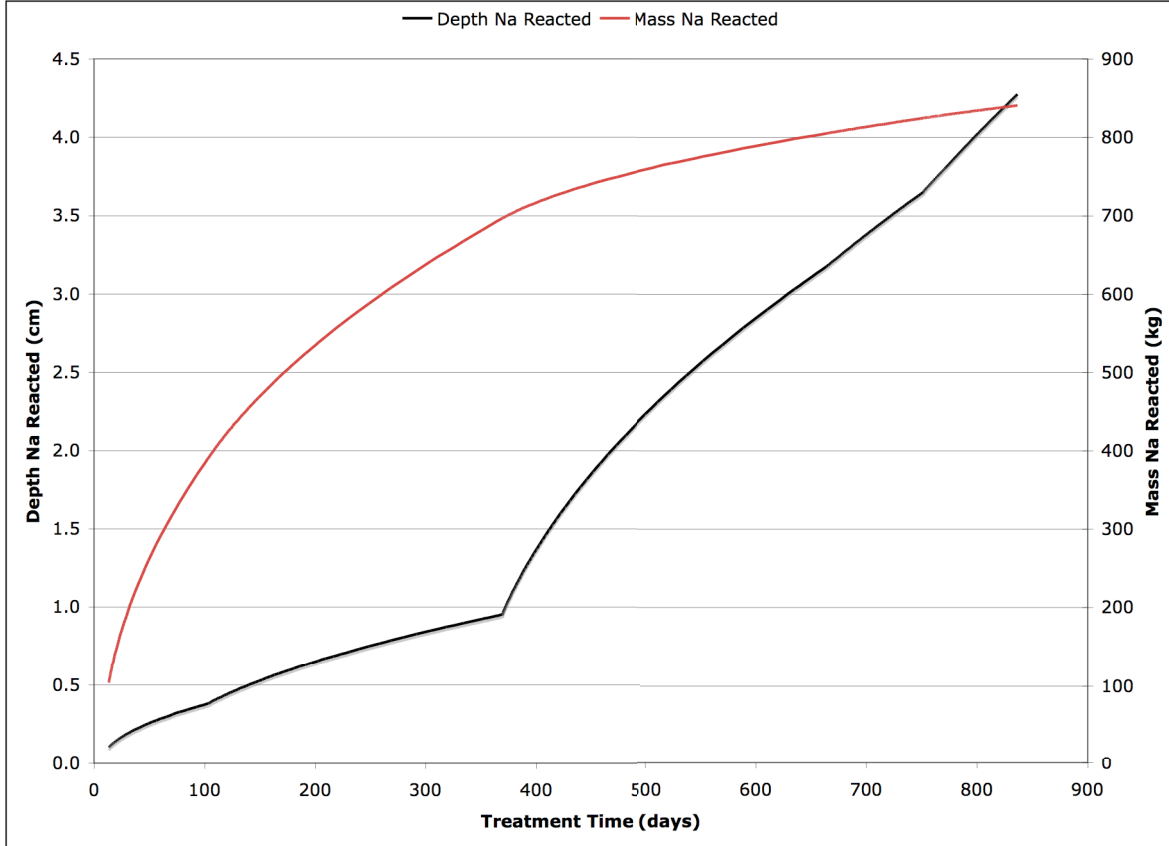


Figure 7. Predicted Na Reaction Depth and Mass versus Treatment Time

### 3.1.3 Resumption and Completion of Carbonation

Carbonation of the EBR-II primary tank was restarted on 18 May 2004 and continued until 8 December 2005 with only brief interruptions for maintenance of the humidification cart. A final purge with dry carbon dioxide was performed between 4 January 2006 and 13 January 2006 in order to remove residual hydrogen from the system. The flow rate of humidified carbon dioxide was maintained at 5 scfm and the water tank on the cart was heated to approximately 35 °C in order to increase the moisture content of the carbon dioxide. Figure 8 shows the measured hydrogen concentration and mass flow rate of the exhaust gas for the treatment period. From the figure it must be noted that a rising hydrogen concentration occurs when the mass flow rate through the exhaust line falls to zero. This is due to stratification of hydrogen within the EBR-II primary tank during a “no-flow” condition, with the lighter hydrogen concentrating at the top of the tank. The stratification disappears when flow is restored, as indicated by the sudden drops in measured hydrogen concentration at about 30 days, 95 days, 240 days, and 590 days in the figure.

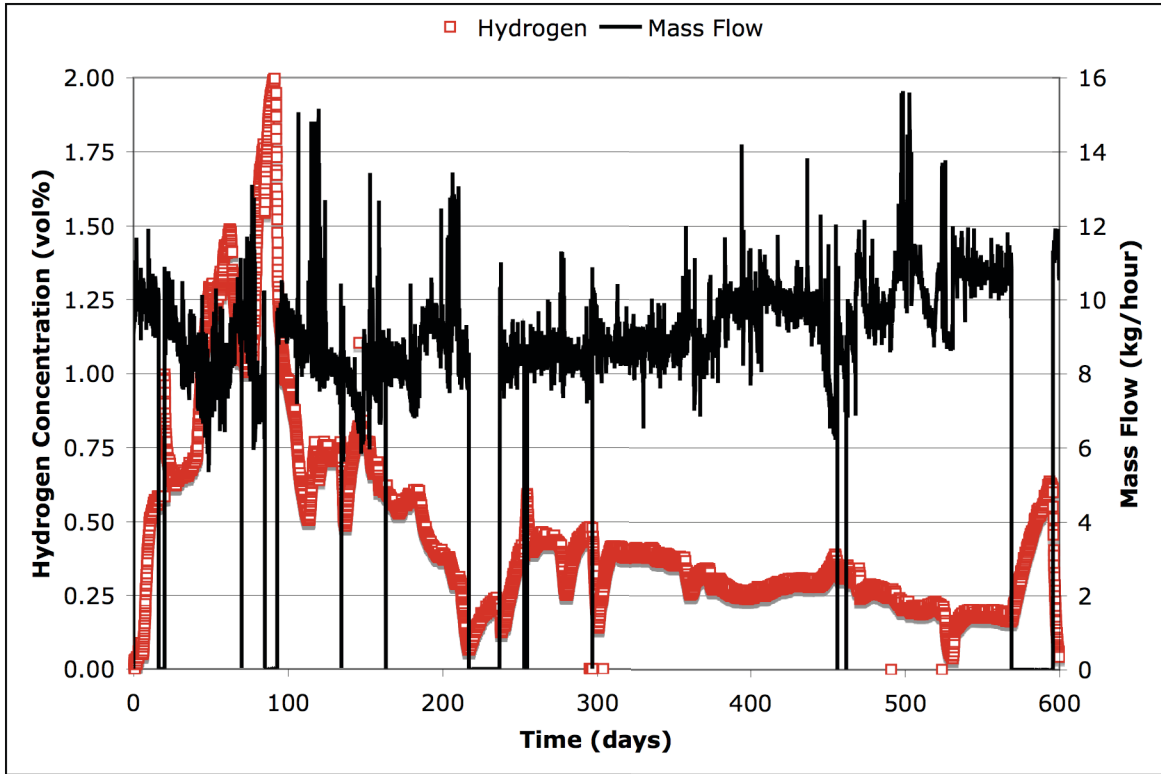


Figure 8. Measured Hydrogen Concentration and Mass Flow After Resumption of Carbonation

The observed reaction rate of moisture with the residual sodium in the primary tank followed the trend observed in Figure 6. Figure 9 shows a comparison of the model (shown previously) with the observed sodium reaction rate in gallons/day.

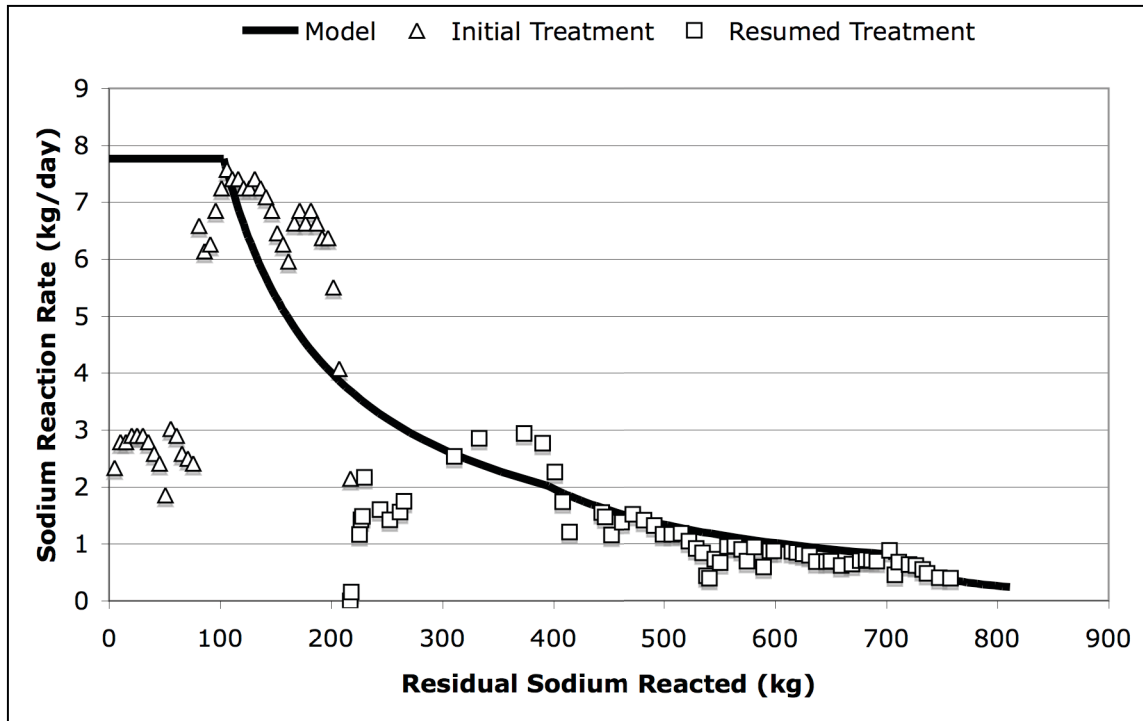


Figure 9. Comparison of Predicted versus Measured Sodium Reaction Rates

Initially, the observed rate was much lower than the modeled rate because the treatment process was operated only intermittently while the model assumed that the treatment process be operating at steady state. When the treatment process was changed to operate on a continuous basis, the observed sodium reaction rates matched more closely to the model. Upon resumption of treatment in May 2004, the observed reaction rates were also below the predicted rate, but eventually rose to correspond to the predicted rates.

Figure 10 shows a comparison of the predicted reaction time for a given volume of residual sodium in the primary tank and the observed reaction time. The observed reaction time is longer than the predicted reaction time. This difference is due to treatment interruptions from routine system maintenance operations (e.g., hydrogen or oxygen meter re-calibration, water tank refills) and occasional variations in the flow rate or humidity content of the carbon dioxide that are not accounted for in the simple reaction model. Over time, treatment delays caused by these interruptions and variations accumulate over time, causing the observations to lag the predicted total reaction time. The trend in the predicted treatment time is consistent with the observed data.

A hydrogen mass balance was used to calculate the mass of sodium reacted in the primary tank. From the data, it is determined that approximately 760 kg or 775 liters of residual sodium were reacted inside the primary tank during carbonation. This amount corresponds to approximately 70% of the original residual sodium. All residual sodium on the exposed sodium surfaces (i.e., bottom, sides, top) of the primary tank has been converted into sodium bicarbonate. Residual sodium remains in the EBR-II primary tank at locations 2, 3, 4, 7, 8, 14, 16, and 21, as listed in Table 3. The depth of sodium reacted is estimated to be 2.3 cm at the deepest point and the depth of the sodium bicarbonate layer is estimated to be at most 11.5 cm thick.



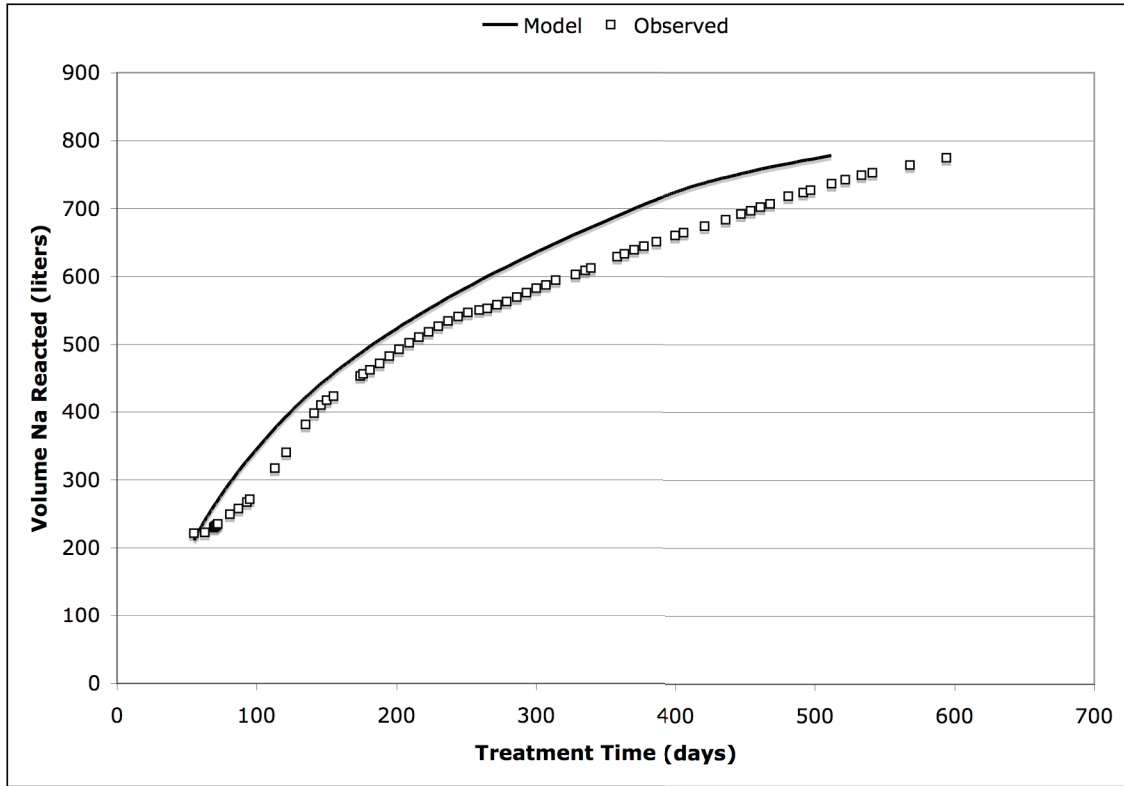


Figure 10. Comparison of Predicted Reaction Time with Observed Reaction Time

### 3.2 Argon System Components

The bulk sodium coolant in the primary tank was covered with an inert gas blanket during reactor operation to prevent sodium contact with air. The cover gas system drew gas from this inert blanket through nozzle penetrations in the primary tank cover. The gas stream was then circulated through the system and returned to the primary tank or vented to the suspect exhaust system. During reactor operation, the primary tank cover gas contained argon, nitrogen sodium vapor, sodium aerosols, and trace levels of other gaseous impurities. The sodium impurities in the cover gas were removed from the gas stream prior to its analysis in on-line monitoring instruments and other system components. The sodium impurities removal was accomplished through the use of two sodium vapor traps operating in series (Vapor Traps #1 and #2). The vapor traps contained a metal mesh which was designed to capture sodium aerosol.

A cover gas cleanup system (CGCS) was used to remove radioactive argon from the primary tank cover gas. The main loop portion of the CGCS consists of a piping arrangement which drew radioactive argon from the primary tank cover gas through a nozzle, passed it through components designed to remove sodium vapor, particulates, and radioactive isotopes, and returned cleaned argon to the cover gas blanket in the primary tank through another nozzle. Part of this system includes the Controlled Temperature Profile (CTP) Condenser.

### 3.2.1 Vapor Traps

Figure 11 shows the relationship between the primary tank, Vapor Trap #1, and Vapor Trap #2.

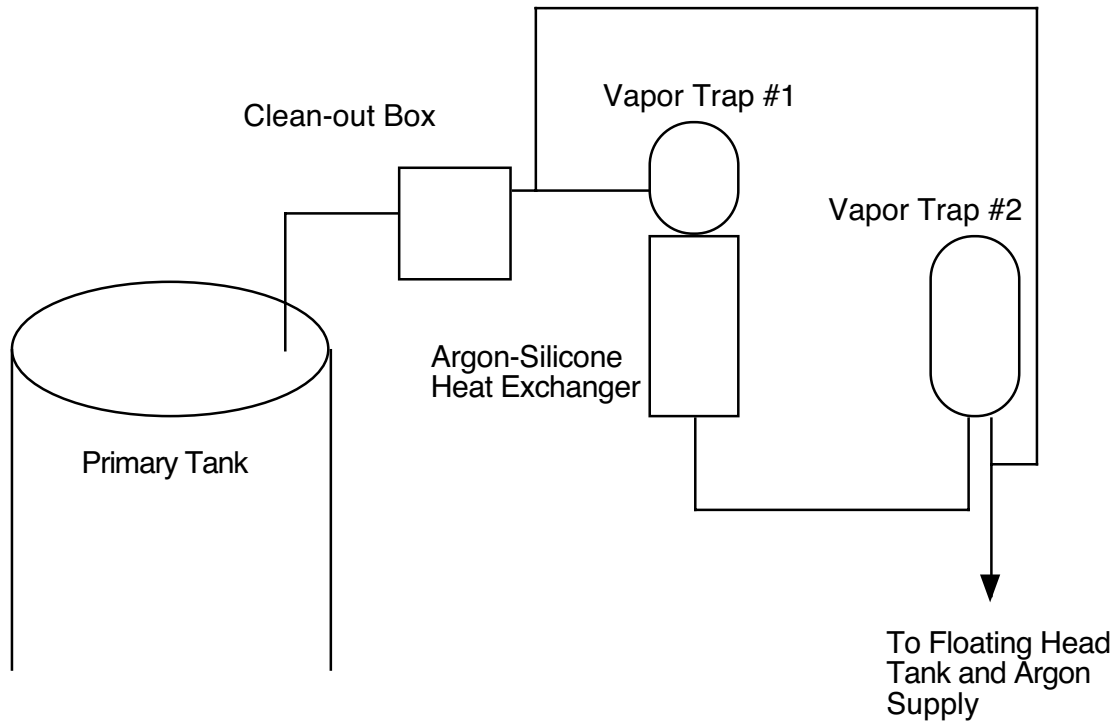


Figure 11. Sketch of Cover Gas Supply System with Vapor Traps

Following the initial carbonation of the primary tank (Section 3.1.1), the vapor traps were treated with humidified carbon dioxide. Humidified carbon dioxide was supplied directly to each vapor trap, and the exhaust gas from the vapor traps were vented through the existing cover gas supply system piping into the primary tank and out the exhaust vent. Venting the exhaust gases through the primary tank was necessary because it was the only location with installed instrumentation.

Vapor Trap #2 was treated first. Carbonation began at 9:30 5 February 2002 and ended 9:00 11 February 2002. The humidification cart was operated with a flow rate of 134 slm and with the water tank heated to approximately 30 °C. Figure 12 shows the measured mass flow rate and hydrogen concentration during the treatment.

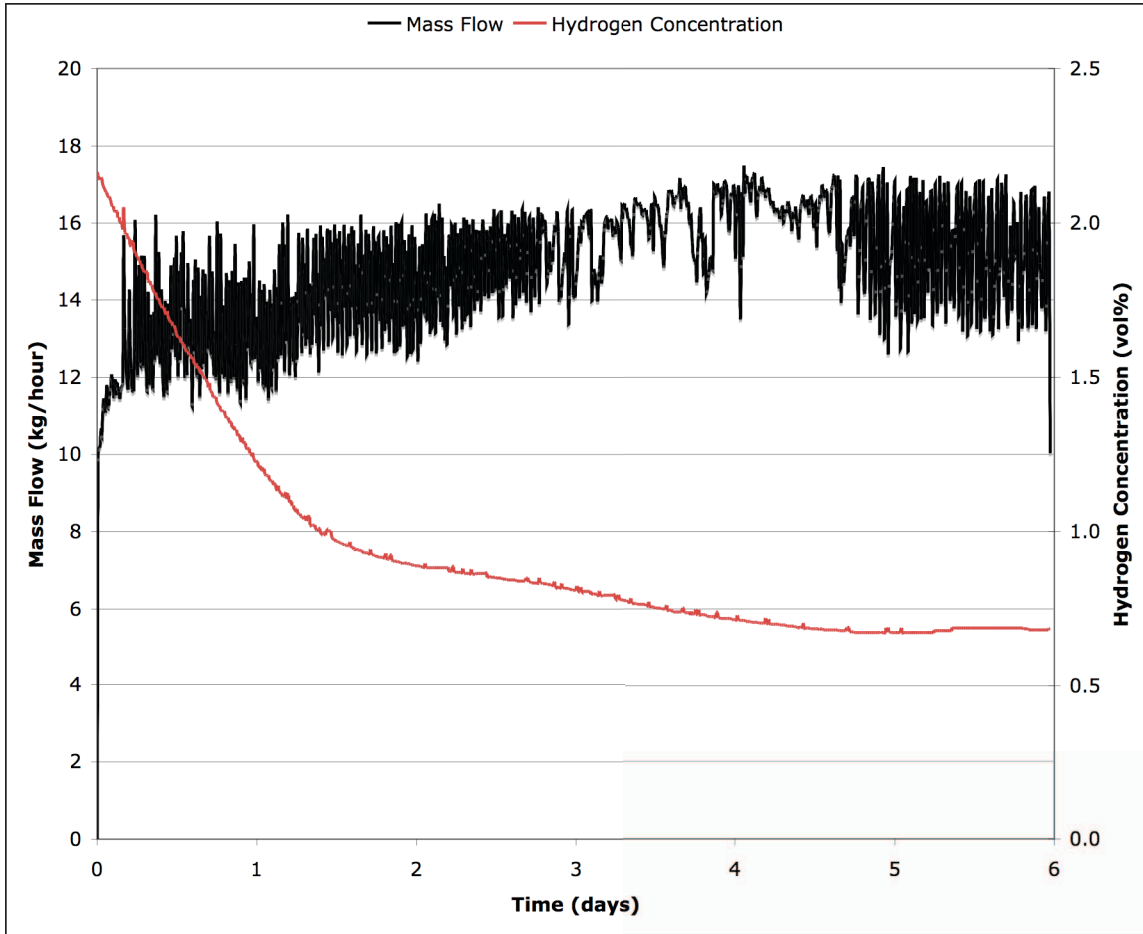


Figure 12. Measured Exhaust Gas Mass Flow Rate and Hydrogen Concentration for Vapor Trap #2.

During treatment of Vapor Trap #2, the measured mass flow rate of exhaust gas was higher than was encountered during initial carbonation of the primary tank. It is believed this higher flow rate was due to supplemental flow of dry carbon dioxide from the floating head tank, which is located adjacent to Vapor Trap #2 in the argon cover gas supply system (see Figure 11). The supplemental flow of carbon dioxide most likely diluted the hydrogen generated from the water-sodium reaction within the vapor trap, resulting in lower measured hydrogen concentrations that would have otherwise been observed.

No moisture was detected in the exhaust gas which indicates that all of the moisture flowing into the vapor trap was reacting with residual sodium somewhere within the system.

No mass balance on hydrogen was performed because prior knowledge indicated that the hydrogen monitor was not calibrated properly. A water balance was used instead to estimate the amount of sodium reacted. Approximately 14 liters of water were evaporated during the treatment of this vapor trap. If all of this water reacted with residual sodium, then a maximum of 17 kg +/- 2 kg of residual sodium would have reacted. No clear end point was reached during this short treatment period, and it cannot be assured that there is no residual sodium remaining inside the vapor trap. The original amount of residual sodium within the vapor trap before treatment was unknown, so no assessment of completion can be made based on the mass balance.

Vapor Trap #1 was treated subsequent to treatment of Vapor Trap #2. Carbonation began at 9:00 11 February 2002 and was stopped at 9:00 18 February 2002. Figure 13 shows the observed exhaust gas mass flow rate and hydrogen concentration during treatment.

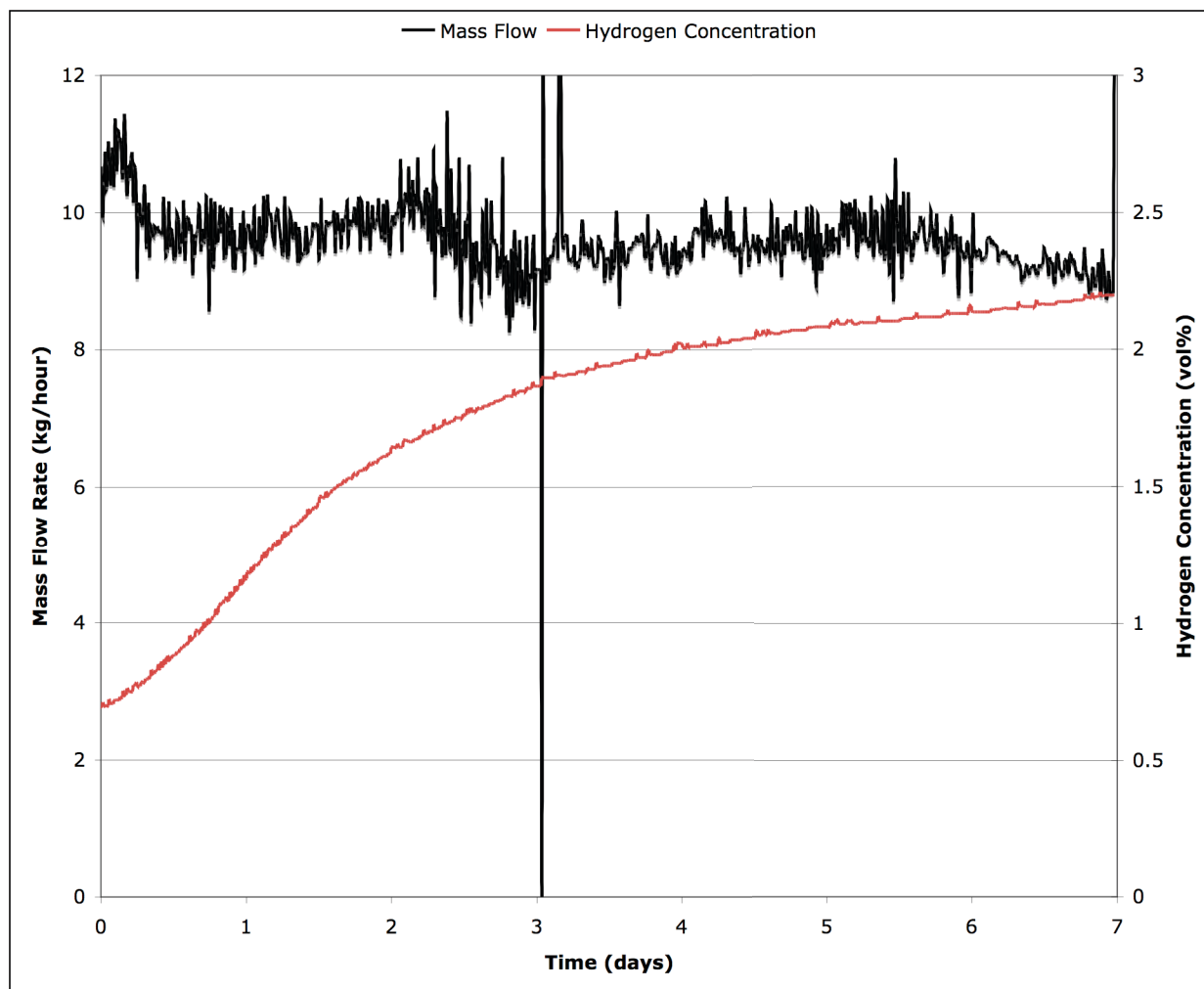


Figure 13. Measured Exhaust Gas Mass Flow Rate and Hydrogen Concentration for Vapor Trap #1.

The observed mass flow rate for Vapor Trap #1 is more consistent with the flow rates observed during the initial carbonation and subsequent treatment of the EBR-II primary tank. The measured hydrogen concentration rose above 2 vol% during this treatment period, which is most likely indicative of the presence of residual sodium within the vapor trap. No end point was reached and it is very likely there is residual sodium remaining within the vapor trap.

No moisture was detected in the exhaust gas, which indicates that all moisture flowing into the vapor trap was reacting with residual sodium somewhere within the system.

Since the hydrogen monitor was suspected of having an offset, a hydrogen mass balance was not used to assess the amount of residual sodium reacted in the vapor trap. Approximately 16 liters of water were evaporated during treatment, and if all of this water reacted with residual sodium, then the amount of sodium reacted would be 15 +/- 2 kg. The original amount of residual sodium within the vapor trap

was unknown, so no comparison between a starting amount and the water mass balance result can be made.

An end point determination for either vapor trap would be difficult at any time given the present configuration of flow paths and instrumentation because the exhaust gases flow through the primary tank before exiting. Any moisture that did not react with sodium in the vapor traps will have emptied into the primary tank and would have been available to react with residual sodium in that vessel. With enough exposure time, eventually it could be assumed the vapor traps were free of residual sodium, but this could probably not be verified without doing a direct visual inspection of the traps.

### 3.2.2 Cover Gas Cleanup System (CGCS)

The cover gas cleanup system, including the controlled temperature profile condenser, was carbonated between 9:00 18 February 2002 and 8:00 26 February 2002. The flow conditions were the same as for the vapor traps: 134 slm with a humidification tank water temperature of approximately 30 °C. Figure 14 shows the measured mass flow rate and hydrogen concentration during the treatment period.

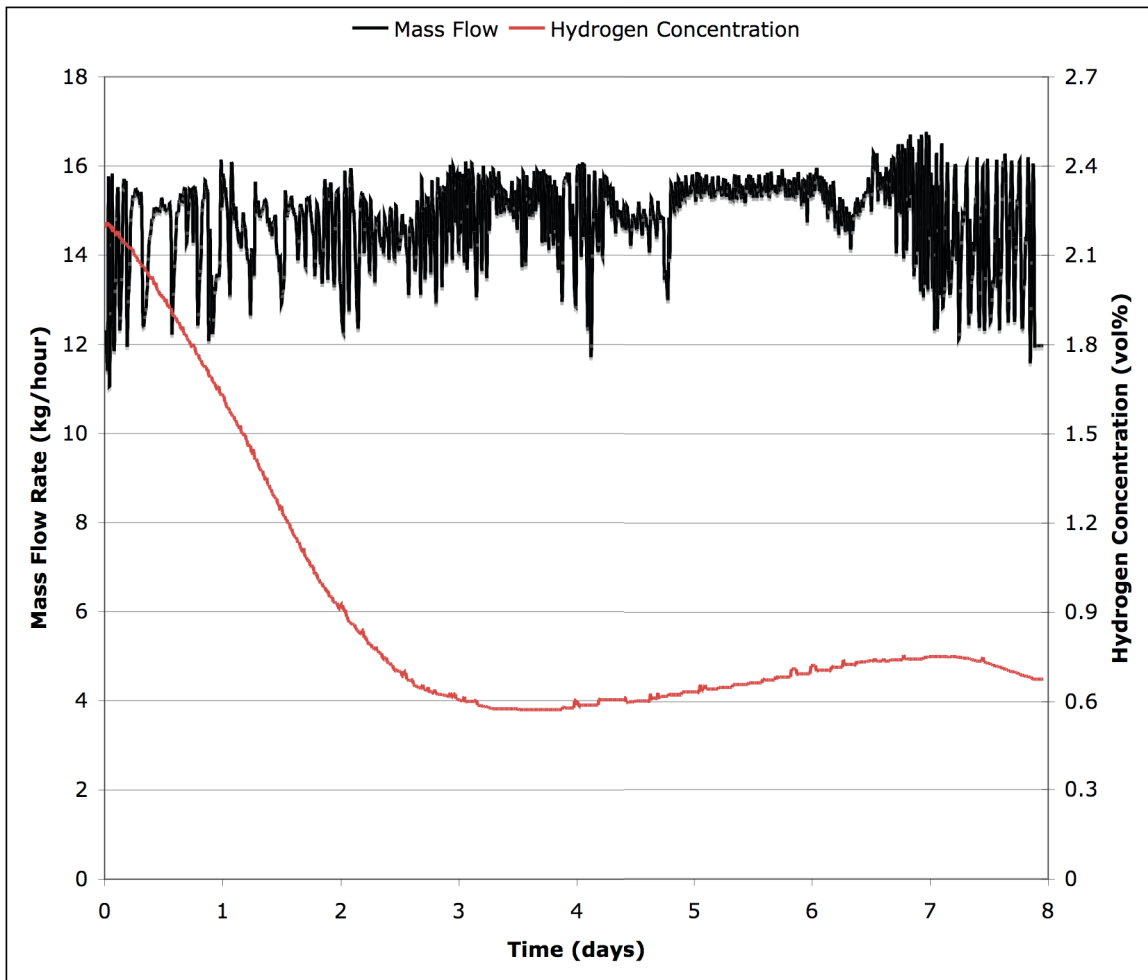


Figure 14. Observed Mass Flow Rate and Hydrogen Concentration During CGCS Treatment

No moisture was detected in the exhaust gas, which indicates that all of the moisture flowing into the cover gas cleanup system was reacting with residual sodium somewhere within the system.

With prior knowledge of an offset in the measured hydrogen concentration, a mass balance on the hydrogen generated was not performed and a water balance was used instead to estimate how much residual sodium had reacted during treatment. Approximately 25 liters of water were evaporated during treatment, which, if all of this water reacted with sodium, would correspond to a maximum of 31 +/- 3 kg of residual sodium. No conclusion could be drawn as to whether all of the residual sodium within the cover gas cleanup system was completely consumed since the treatment gas flows through the primary tank before being exhausted. A visual inspection will need to be performed on the system in order to detect any remaining residual sodium. Also, the original amount of residual sodium within the cover gas cleanup system is unknown, and no comparison can be drawn between the starting amount of sodium and the amount that may have reacted based on the water mass balance.

No further treatment of the vapor traps was performed beyond the completion of the EBR-II Plant Closure Project.

## 4. INTERMEDIATE HEAT EXCHANGER

During the EBR-II Plant Closure Project, the EBR-II intermediate heat exchanger (IHX) was drained of bulk sodium and isolated from the EBR-II secondary sodium system by cutting the inlet and outlet lines to the secondary sodium system and sealing the connections. After sealing the connections, the inlet line for the IHX was modified so that it could be connected to the humidification cart. The outlet was modified also, so that the exhaust gases were vented outside of the building after passing through dedicated hydrogen and oxygen monitors within the EBR-II facility. No mass flow or moisture monitoring was performed of exhaust gases from the IHX.

It is known from engineering drawings and an assessment of bulk sodium removed that almost all of the residual sodium within the IHX is pooled at the bottom of the heat exchanger below the tube sheet. The bottom of the heat exchanger is hemispherical and it is assumed that that residual sodium completely fills the hemisphere. See Figure 15 for an external image of the IHX.

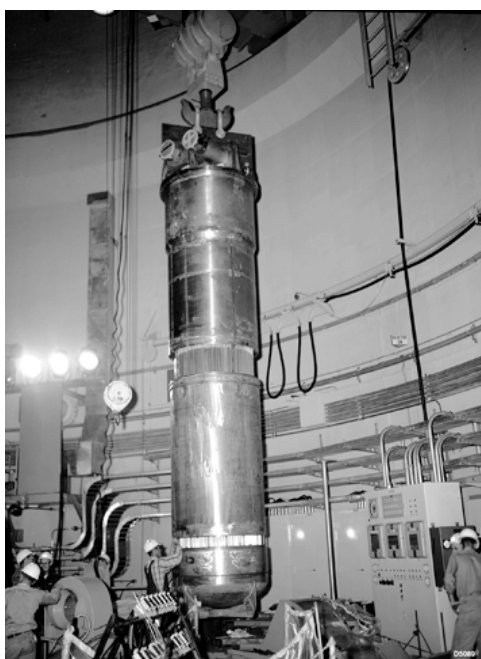


Figure 15. Intermediate Heat Exchanger Before Insertion into EBR-II Vessel

The diameter of the exposed surface of residual sodium at the bottom of the IHX is assumed to be 137.2 cm and equal to the inner diameter of the heat exchanger shell. Other residual sodium may coat the vertical surfaces of the IHX, but the thickness of the sodium on vertical surfaces is believed to be no thicker than 0.2 cm based on visual evidence from the EBR-II primary tank and probably does not comprise a significant percentage of the total amount of residual sodium remaining within the IHX.

Initially, the IHX contained an argon atmosphere, which needed to be replaced with carbon dioxide before carbonation could take place. On 30 August 2001, the cover gas source for the IHX was changed from argon to carbon dioxide though no active flush of the system took place at that time. Prior to the changeover, the IHX was maintained under a static blanket of argon. Then, at 9:10 16 October 2001 a dry carbon dioxide flow rate of 134 slm was established and maintained until 9:55 24 October 2001 to purge any remaining argon from the IHX. No measurements were taken to verify that all of the

argon had been purged, but the flow of carbon dioxide was maintained for a sufficient period of time to ensure that more than 99.9% of the original argon was removed, assuming the system was well-mixed.

Carbonation of the system began at 9:55 24 October 2001 and continued until 15:00 2 November 2001. The humidification cart conditions were 134 slm and a water tank temperature between 18 and 24 °C (ambient temperature conditions). The measured hydrogen concentration during the treatment period is shown in Figure 16. There is no good explanation for the observed fluctuations in the measured hydrogen concentration aside from the observation that the temperature in the treatment area varied on a 24-hour cycle. The heat in the humidification cart water tank was not used and the temperature of the water could float with the room temperature. The variations in temperature may have caused the water content in the carbon dioxide to vary with time, leading to the observed periodic fluctuations in the observed hydrogen concentration.

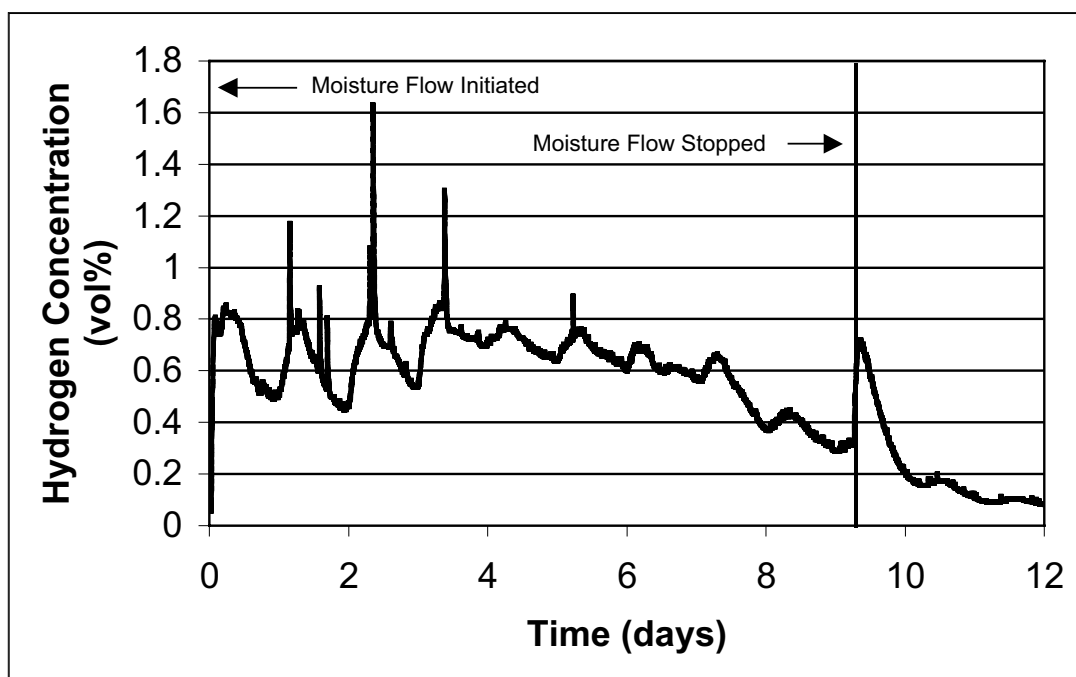


Figure 16. Measured Hydrogen Concentration During Carbonation of IHX

Though no flow meter was installed on the exhaust, an integration of the measured hydrogen data can be performed to get an estimate of the amount of residual sodium reacted. If it is assumed that the flow rate of gas is 134 slm, the atmospheric pressure is 0.83 atm, the inlet temperature of the gas is 16 °C, and the gas volume within the IHX is 512 liters, then the concentration data can be integrated to generate an estimate that 20 kg of sodium reacted during the carbonation process.

During carbonation, approximately 17 liters of water were evaporated from the humidification cart water tank. If all of this water reacted with residual sodium, then the water would have consumed 21 kg +/- 1 kg of sodium. This number is consistent with the estimate obtained from an integration of the measured hydrogen concentration data.

The decision was made to stop carbonation of the IHX after it was decided that the sodium bicarbonate layer on the exposed sodium surface at the bottom of the vessel would eventually grow to the



point where it might block the flow paths through the tube sheet, thus preventing the use of further carbonation or other gas-based in-situ treatment method.

No further treatment of the IHX was performed beyond the completion of the EBR-II Plant Closure Project.



## 5. FUTURE TREATMENT OPTIONS FOR EBR-II

Carbonation of residual sodium within the EBR-II facility has allowed for treatment of residual sodium with minimum expenditure and a low level of process surveillance. No safety problems occurred during treatment, and no unsafe hydrogen conditions occurred at any time. The treatment process operated in a predictable manner and was used successfully to react approximately 70% of the residual sodium in the EBR-II primary tank, approximately 50% of the residual sodium within the EBR-II secondary sodium system, and smaller amounts in related EBR-II systems (i.e., IHX, cover gas systems). The carbonation process has proven to be very useful for reacting residual sodium in shallow pools and on vertical surfaces and has served its role well in reducing the inventory and exposed surface area of the remaining residual sodium deposits in the EBR-II systems.

At this point in the treatment process, however, carbonation has become less effective and slower, and other treatment methods are needed to completely react, treat, or remove the remaining residual sodium. The carbonation treatment process is now at a point of diminishing returns, and faster, more aggressive treatment methods are needed to carry the work to completion. The future treatment methods and sequences are described in this section.

### 5.1 EBR-II Primary Tank

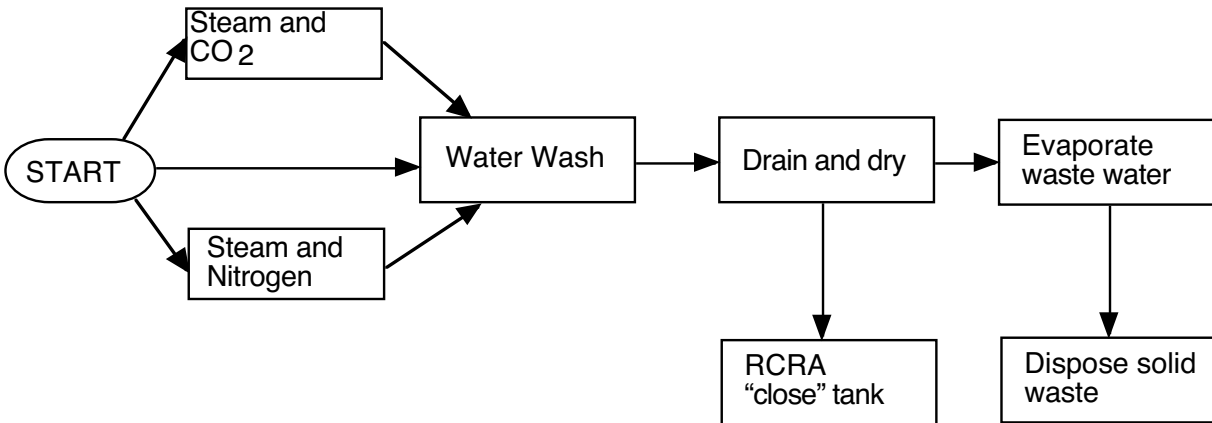


Figure 17. Future Treatment Path of EBR-II Primary Tank

With the decline in sodium reaction rate with the carbonation method, more aggressive techniques are needed to react the residual sodium remaining in the EBR-II primary tank and to achieve RCRA “clean closure.” A more aggressive treatment path is shown in Figure 17. First, steam might be employed to react still further the remaining residual sodium. This would have the effect of greatly increasing the moisture content of the gas and increase the moisture gradient across the established sodium bicarbonate/carbonate layer. Increased sodium reaction rates would result. It would also push the humidity content to above the saturation point and lead to the creation of fog and liquid condensation inside the tank, which would also react readily with any sodium metal with which it comes into contact. Steam would also be able to penetrate more deeply into areas above the liquid fill line in the primary tank than humidified carbon dioxide and react more residual sodium in those places.

There may be some performance and safety implications in choosing either carbon dioxide or nitrogen as the steam carrier gas. If carbon dioxide is used, no pools of liquid sodium hydroxide would form because of the chemical reaction between NaOH and CO<sub>2</sub>, and the probability of large uncontrolled sodium-water reactions occurring would be reduced. A disadvantage of this approach may be the formation of harder, denser layers of sodium carbonate on top of the residual sodium deposits, which may limit the extent of reaction. Using nitrogen as a carrier gas would prevent the formation of thicker sodium carbonate layers and may allow for deeper penetration of the steam, but may also lead to the formation of pools of liquid sodium hydroxide and increased risk of large reaction excursions. Given the current environmental permit that governs treatment of EBR-II, a change in cover gas from carbon dioxide to nitrogen would require a lead time of six months to alter the existing permit while no change is required if steam is injected using a carbon dioxide carrier gas. An analysis still needs to be performed to estimate the additional amount of residual sodium that might be reacted when using nitrogen or carbon dioxide and whether the application of a steam treatment step will be effective in reducing the safety risks in the subsequent water wash step.

Regardless of whether a steam treatment step is used, the primary tank will be water washed. This would entail filling the primary tank with water and allowing the liquid water to dissolve the carbonate layers and react with the remaining residual sodium. The water fill operation would be performed in stages in response to the degree of hydrogen gas generated and the estimated locations of residual sodium pockets (see Table 1). Fresh water might be used to perform the water wash, or wash water from treatment of the EBR-II secondary sodium system might be recycled for this purpose. It is anticipated that water may need to be re-circulated inside the primary tank as the water wash is being performed, and that perhaps some wash water treatment may need to be performed concurrently with the water wash operations in order to remove some of the carbonate/hydroxide load in the wash water. It is expected that the wash water will have a high pH but should be buffered somewhat by the sodium bicarbonate and carbonate already present. It is anticipated that a single fill will be enough to react all of the residual sodium and dissolve all of the reaction products. When the bulk water is drained, a residual amount of water (and dissolved reaction by-products) will remain in the primary tank.

One advantage of keeping carbon dioxide as the cover gas during any steam treatment step becomes apparent once the water wash step commences. Having a carbon dioxide cover gas during the water wash step would help lower the pH of the wash water as the water wash step is performed. Dissolved carbon dioxide in the wash water would be available to react with hydroxide ions in the wash water to form carbonate and bicarbonate ions. This would reduce the corrosion hazard of the wash water and lower the hazard to workers and treatment equipment as a result.

The bulk liquid withdrawn from the primary tank may be neutralized by bubbling carbon dioxide through it if necessary, and then evaporated to recover the dissolved materials. The water treatment process has not yet been designed, but it may involve the use of evaporators, forced circulation crystallizers, filters and solid dryers. Water treatment will be necessary because the full volume of liquid waste would be prohibitively expensive to dispose directly, and the amount of solid material that could be recovered from the wastewater has a much smaller volume than the original liquid. From past experience working with sodium carbonate solutions on other projects, the use of a thin film evaporator for the evaporation task is not recommended due to the difficulty in maintaining sustained operation of the evaporator.

Once the bulk liquid has been drained from the primary tank, the primary tank's external and internal heaters would be activated to heat the tank and boil away the residual moisture. In addition, the primary tank might also be flushed with dry air to remove the excess moisture.

After these treatment steps, the EBR-II primary tank will be dry, clear of residual sodium, and may contain small amounts of dried sodium carbonates and perhaps some sodium hydroxide. Under the existing U.S. Resource Conservation and Recovery Act (RCRA) regulations, the tank will be considered RCRA empty and will be “clean closed.” The tank will be open to the atmosphere.

## **5.2 Primary Tank Cover Gas Systems**

These systems include the primary tank cover gas supply system and the cover gas clean-up system.

Carbonation treatment of the Vapor Traps #1 and #2 will be resumed to react any remaining sodium aerosol in the traps. Success will be indicated by low hydrogen concentration in the primary tank exhaust gas. At the end of carbonation of the primary tank, the measured hydrogen concentration in the exhaust was below 0.25 vol% (see Figure 8). The measured hydrogen concentration for both vapor traps during the EBR-II Plant Closure Project was above this limit (see Section 3.2.1), which indicates that residual sodium still remains inside the vapor traps. Treatment of the vapor traps with humidified carbon dioxide will be performed before the primary tank is treated with steam or water washed.

The primary tank cover gas supply system will remain in-place until the primary tank has been water washed. Afterward, the system will be removed. The system components will be disassembled and inspected for residual sodium. If any residual sodium is found, the affected components will be sent to the Sodium Component Maintenance Shop for treatment and disposal. The oil in the floating head tank will be drained and disposed separately.

Carbonation treatment of the cover gas clean-up system might also be continued, since no clear end-point was reached in the treatment of this system during the EBR-II Plant Closure Project (see Figure 14). If carbonation is re-started, humidified carbon dioxide will be supplied to the system until the measured hydrogen concentration falls below 0.25 vol%. After reaching this threshold, the components of the cover gas clean-up system will be removed and treated at the Sodium Component Maintenance Shop. The lead shielding used the system will be removed as it becomes unnecessary and disposed or recycled. Charcoal in the charcoal absorbers will be removed and disposed. If carbonation of the cover gas clean-up system is re-started, it must be performed before the cover gas inside the primary tank is changed from carbon dioxide to nitrogen or air. After completing carbonation, the components of the system can be removed at any time as funding permits.

## **5.3 Intermediate Heat Exchanger**

No further carbonation treatment of the intermediate heat exchanger (IHX) will be performed due to the internal space limitations of the IHX. The free volume within the IHX would not be sufficient to contain the sodium bicarbonate created by the sodium/water/carbon dioxide reactions if all of the residual sodium within the IHX were to be reacted.

After the EBR-II primary tank has been water washed, the IHX will be pulled from the primary tank in the same manner it was inserted during construction (see Figure 15) and transported to the Sodium Component Maintenance Shop for treatment. An existing procedure for performing this operation will be used and will be modified as needed to address the changed conditions of the primary tank. The existing primary tank heaters might be employed to heat the primary tank if necessary to remove the IHX. The bottom of the IHX will be tapped with a drainage hole and the sodium contained within it will be heated to melt and remove it. Then, the IHX will be cut apart with cutting saws and/or plasma torches, and the pieces will be water washed in the Sodium Component Maintenance Shop's Water Wash Vessel. After

washing, the pieces will be disposed in approved drums and waste boxes at an INL-approved waste disposal site.

The hole created in the primary tank cover by the removal of the IHX will be covered with a metal plate.

If the IHX cannot be removed, the IHX would be treated in place, though the risks of uncontrolled sodium-water reactions would be much greater. For in-place treatment, the IHX would be flushed with steam-and-nitrogen until hydrogen is no longer generated. Then the IHX would be flushed with water until the pH of the flush water begins to approach neutrality. The flush water would then be treated using the same equipment as was used to treat the wash water for the primary tank. The steam treatment and water wash systems would need to be designed to handle pressure excursions in order to avoid damage to personnel and equipment.

## 5.4 Other EBR-II Systems

The EBR-II systems highlighted here are part of a larger list of EBR-II systems that will need treatment in order to achieve “clean” status. The total list of systems is given below.

1. Primary sodium tank
2. Primary tank cover gas system
3. NaK transmitters
4. Secondary sodium purification, gas recirculation, and HMLD systems
5. Secondary sodium system and primary sodium transfer line
6. Intermediate heat exchanger and reactor building piping
7. Rotating plugs, seals, and superstructure components
8. Primary tank heaters
9. Primary tank nozzles
10. Fuel handling systems
11. Primary tank cover gas sampling supply system
12. Shutdown coolers
13. Reactor building storage pit and manipulators
14. Cover gas cleanup system
15. Alcohol recovery station (SCMS)
16. Primary purification and sampling support system
17. Radioactive sodium chemistry loop
18. Reactor building storage holes
19. Argon purge system
20. Argon cooling system
21. Hydrocarbon analyzers
22. Failed fuel transfer and gas collection system
23. Fuel element rupture detection system
24. Secondary sodium recirculation system

Multiple timelines for the performance of these tasks have been developed, and these time lines show that the clean-up tasks would require between 6 to 12 years to accomplish, depending upon the rate of work and availability of funding. The clean-up effort might be accelerated still further, but changes would need to be made to the existing Sodium Component Maintenance Shop to allow for parallel processing of waste materials.

A Rough Order of Magnitude estimate of how much it might cost to do all of the clean-up tasks has been generated. This estimate shows that it may take approximately \$25M (FY 2006 dollars, unescalated) to perform all of the clean-up tasks. This estimate will be revised and made more accurate as the individual planning is performed for each individual task.

## REFERENCES

1. INL/EXT-05-00280, Version 0, Technical Information on the Carbonation of the EBR-II Reactor, Summary Report Part 1: Laboratory Experiments and Application to EBR-II Secondary Sodium System, S.R. Sherman, April 2005.
2. U.S. Patent 6,919,061, “In-sit Method for Treating Residual Sodium”, July 19, 2005, S.R. Sherman, S.P. Henslee, assigned to USA as represented by the U.S. Department of Energy.