10-5-2006

An Assessment of Potential Pathways for Release of Gaseous Radioactivity Following Fuel Damage During Run 14 at the Sodium Reactor Experiment

David A. Lochbaum

Santa Susana Field Laboratory

AN ASSESSMENT OF POTENTIAL PATHWAYS FOR RELEASE OF GASEOUS RADIOACTIVITY FOLLOWING FUEL DAMAGE DURING RUN 14 AT THE SODIUM REACTOR EXPERIMENT

BY

DAVID A. LOCHBAUM

PREPARED FOR THE
SANTA SUSANA FIELD LABORATORY
ADVISORY PANEL

5 OCTOBER 2006

Note: The author is the Nuclear Safety Engineer for the Union of Concerned Scientists. However, this assessment was conducted during evenings and weekends and reflects the opinions and positions of the author, not the UCS.
This work was conducted for the SSFL Advisory Panel, a project of the Tides Center. Although the author performed this work without compensation, the work of the SSFL Panel was supported by funds provided by the California State Legislature through a contract from the California Environmental Protection Agency and a grant from the Citizens Monitoring and Technical Assistance Fund. The statements and conclusions expressed in this report are those of the author and do not necessarily represent those of the Tides Center, the CMTA Fund, or CAL-EPA. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as actual or implied endorsement of such products.

EXECUTIVE SUMMARY

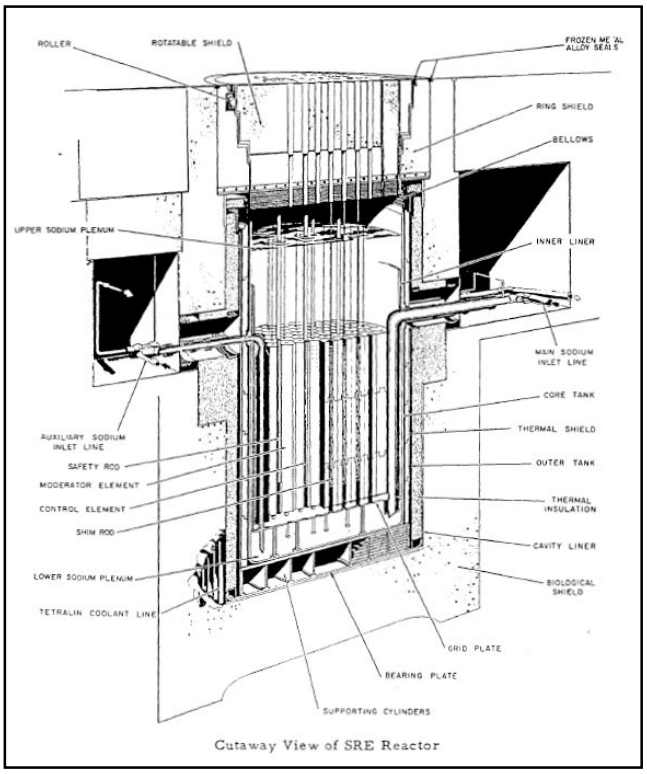
The Sodium Reactor Experiment (SRE) experienced extensive fuel damage during power run 14 in July 1959. Thirteen of forty-three fuel elements in the SRE reactor core failed due to overheating when the cooling flow provided by liquid sodium was blocked or partially blocked by tetralin that had leaked into the primary sodium loop during prior power runs. Fission products were released from the damaged fuel into the primary sodium loop. Some of the fission products leaked from the primary sodium loop into the high bay area, a region inside the building housing the SRE reactor. Other fission products flowed with the helium cover gas over the liquid sodium in the reactor pool to gaseous storage tanks. Fission products from the high bay area and from the gaseous storage tanks were processed through the filters of a ventilation system and discharged to the atmosphere.

Scant and disconnected data prevented a quantitative assessment of what got out when. A qualitative assessment, relying on experience from the subsequent fuel damage experienced at another sodium-cooled reactor, Fermi-I, and on results from experimental studies was performed to estimate the amount of radioiodine and cesium reaching the environment following the July 1959 accident at SRE.

Based upon the fraction of the reactor core damaged (30 percent), the analytical value for fraction of radioiodine and cesium release from damaged fuel (10 percent), and an empirical value for effectiveness of the hold-up and filter performance of the ventilation system (10 percent), it was concluded that the fraction of the total cesium inventory within the SRE reactor core at the time of the July 1959 accident reaching the environment ranged from 0.3 percent to 30 percent and the associated release fraction for radioiodine ranged ranged between 3 percent and 30 percent. Balancing factors which drive the release fraction towards the upper bound and factors driving the release fractions towards the lower bounds yields the conclusion that it is reasonable to assume a release fraction of 15 percent is likely closer to the actual release fraction than is either end point.

SRE DESIGN

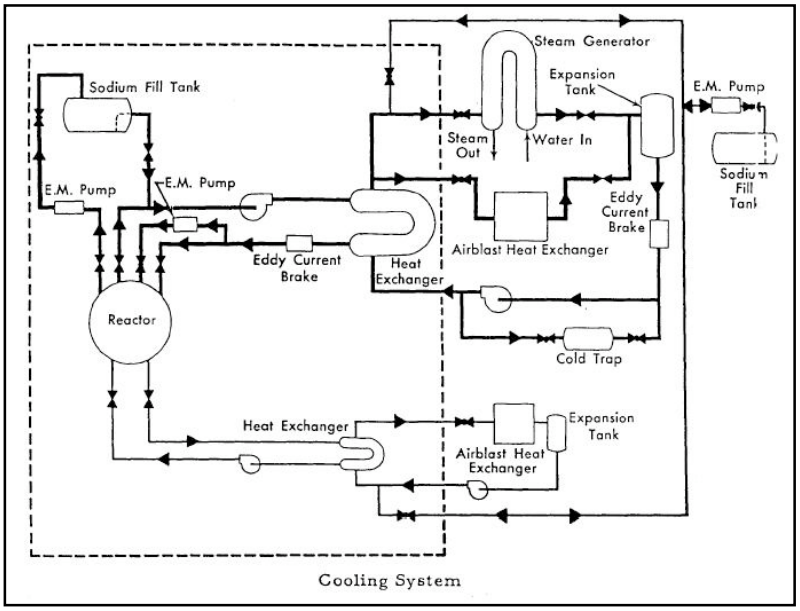
The Sodium Reactor Experiment (SRE) featured a sodium-cooled, graphite-moderated nuclear reactor. The reactor core sat in the lower portion of a vessel lined with stainless steel and filled with liquid sodium.[1] The SRE reactor core contained forty-three (43) fuel elements, each comprised of seven (7) fuel rods. A fuel rod was a stainless steel tubes six feet long filled with uranium fuel slugs.



Cutaway View of SRE Reactor

At full power, sodium at a temperature of approximately 500°F passed through plenum area beneath the reactor core, flowed upward through the fuel channels absorbing heat released from the fuel elements, and discharged into the upper pool about 6 feet deep above the core at an average temperature of nearly 950°F. The space above the surface of the liquid sodium was filled with helium gas maintained at a pressure of approximately three pounds per square inch.[2]

Piping circulated 50,000 pounds of heated liquid sodium from the reactor vessel to either of two available heat exchangers. As shown in the lower portion of the figure, one option transferred the heat from the primary sodium to a secondary loop which in turn dissipated the heat to the atmosphere via an airblast heat exchanger. The second option, shown in the upper portion of the figure, transferred the heat from the primary sodium to a secondary sodium loop which in turn dissipated the heat in a steam generator that boiled water to make steam for use in a turbine/generator that produced electricity for the Southern California Edison grid.[3]



Cooling System

[1] ACRS - 1958
[2] AI - 1959
[3] AI - 1959

RUN 14

As implied by its name, the primary purpose of SRE was not to generate electricity but rather to test the performance of the sodium-cooled, graphite-moderate reactor design, fuel element designs, and the performance of materials. The reactor would be started up and operated for a period, then shut down for examinations and/or to reconfigure test conditions. These periods of reactor operation were termed power runs.

During run 14, the thermocouples that monitored the temperatures at the exit of individual fuel channels in the reactor core and plotted them continuously on a recorder in the control room showed large (greater than 100ºF) differences. Some differences were to be expected due to variances in fuel element power levels and fuel channel cooling flow rates, but the magnitude of the observed differences could not be explained.

On July 13, 1959, the as-yet undetected problem causing the temperature differences manifested itself in another way. Quoting from the subsequence investigation reports:

> [After the reactor restarted on July 13, 1959] *The moderator temperature did not respond properly to an increase in sodium flow. It appeared that very little sodium was leaking across the grid plate for moderator coolant.*
>
> and
>
> *At this time* [1821 on July 13, 1959] *the power started to increase more rapidly, so control rod insertion was started. In spite of this rod insertion the rate of power rise continued to increase. Three positive period transients indicating about a 50-sec period were observed at about 1824 and at 1825 a 7 _ sec period was indicated. At this time the reactor power was rising rapidly; so the reactor was scrammed manually by the operator.*[4]

The temperature anomalies were now accompanied by uncontrolled reactivity transients that caused the power level of the reactor core to rise. The power transients experienced on July 13th resulted in the SRE power level increasing from 4 megawatts to about 14 megawatts. The 7 _ second period observed at 1825 (6:25 pm) meant that the reactor power level increased by a factor of 2.718 in less than eight seconds. In other words, the power level was doubling in less than eight seconds. Left unchecked, this rate meant a reactor operating a 1 percent of its licensed limit would reach 2 percent in 8 seconds, 4 percent in 16 seconds, 8 percent in 24 second, 16 percent in 32 seconds, 32 percent in 40 seconds, 64 percent in 48 second, and 128 percent in 56 seconds.

Attempts were made to remedy the temperature problem. Examinations made after prior runs revealed some debris in the liquid sodium that might obstruct cooling flow past the fuel elements. Consequently:

> *Between 0000 and 0800, on July 24, while jiggling elements in an attempt to dislodge foreign material and hence lower the fuel-channel outlet temperatures, it was noted that the elements in core channels 10, 12, 35, and 76 were stuck in place.*[5]

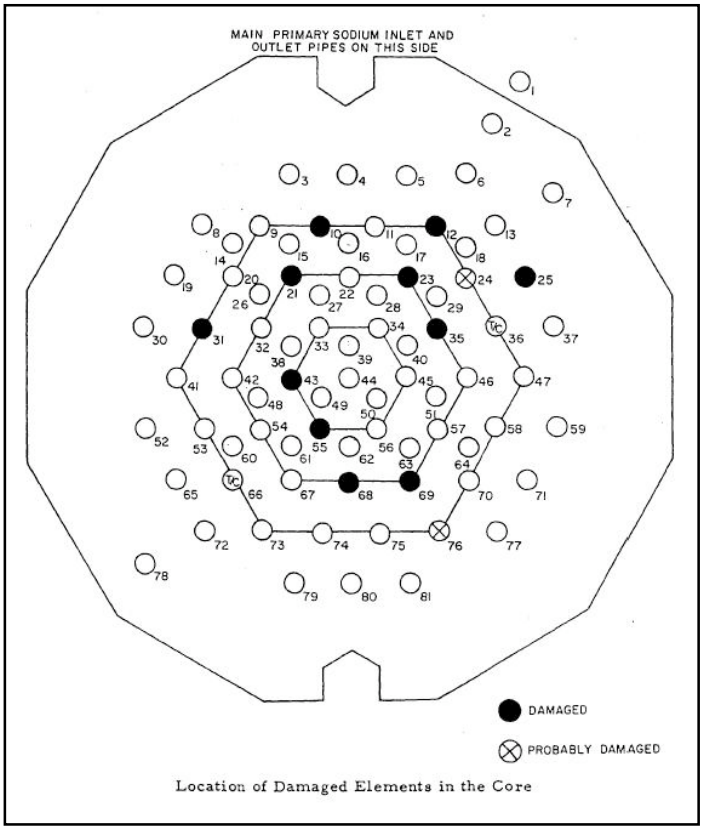
---

[4] AI - 1959
[5] AI - 1959

No recorded attempts were made to dislodge these fuel elements before SRE was restarted. Reactor operation was not resumed for long:

> *On July 26 at 1120, the reactor was shut down after a total exposure of about 16 Mwd on run 14. The shutdown was made to examine each fuel element which had been running hot with the television camera and to try to clear any obstructions in channels. The first damaged fuel element was observed at 1915 on July 26.*[6]

Thirteen (13) of the forty-three (43) fuel elements in the SRE reactor core had been damaged. The locations of the damaged fuel elements are indicated in the figure. All four of the fuel elements (10, 12, 35 and 76) found to be stuck on July 24th were later found to be damaged.



Location of Damaged Elements in the Core

After examining the damaged fuel elements removed from the SRE reactor core, the cause of their failure was determined:

> *The fuel element cladding failed when the stainless steel alloyed with the uranium fuel. Insufficient coolant flow in the fuel channel permitted the temperature of the fuel-clad interface to increase to a level where solid-solid diffusion of iron and uranium could occur at a significant rate.*
>
> and
>
> *The fuel cladding failed as a result of the formation of low-melting iron-uranium alloy which was produced because of partial blockage of some of the coolant passages and local overheating of the fuel elements. Coolant channel blockage was initiated by accumulation of the decomposition products of tetralin which had leaked into the primary system. Sodium oxide and sodium hydride may have contributed to this situation.*[7]

> *It is clear that still higher temperatures were reached in the region of failure, and it is likely that sodium boiling occurred in that region.*[8]

> *Severe overheating of some of the fuel elements is known to have existed. Some melting of uranium in the Fe-U eutectic formation also occurred.*[9]

---

[6] AI - 1959
[7] AI - 1959
[8] AI – 1961a
[9] AI - 1962

Tetralin had been used instead of water to cool some components, like the bearings for the sodium pumps. During prior power runs, some of the tetralin had leaked into the liquid sodium of the primary loop. Apparently, that tetralin "congealed" within the liquid sodium and blocked cooling flow through many of the fuel elements. Varying degree of flow blockage accounted for the temperature differences measured at the outlets of the fuel channels. Flow blockage also accounted for the power transient experienced on July 13th.

This evidence clearly demonstrates the first containment barrier between fission products produced by the nuclear fuel during reactor operation and the environment had been breached for 13 of the 43 fuel elements during run 14.

RADIOLOGICAL CONSEQUENCES

The company that operated SRE reported:

> *In spite of the cladding failure to at least 11 of the fuel elements, no radiological hazard was present to the reactor environs.*[10]
>
> and
>
> *In spite of the cladding failure to 13 fuel elements and the release to the primary coolant of several thousands of curies of fission product activity, no radiological hazard was presented to the reactor environs.*[11]

The validity of this conclusion is not intuitively obvious given that about one third (13 of 43 fuel elements) of the SRE reactor core was severely damaged and the reactor continued to operate for nearly two weeks after this damage likely occurred (July 13, 1959). For this conclusion to be valid, the "several thousands of curies of fission product activity" released to the liquid sodium in the primary loop would have had to be contained such that the amount released to the environs did not pose a radiological hazard. This assessment looked at the amount of radioactivity reported in the primary loop and SRE facility and the potential for pathways to comprise the asserted "containment."

A chronological listing of radioactivity measurements at SRE before, during, and after the core damage event:

- *On May 26* [1959], *the core gas radioactivity was found to be 1.7 x 10^{-3} _c/cm^3 activity was assumed to be xenon-133 and was considered to be normal. Xenon activity had been observed after reactor operation for many months and was attributed to small pin-hole leaks in the cladding of a few elements or to uranium contamination from the outside of new fuel elements.*[12]

- *The reactor cover-gas radioactivity was measured on June 13, 16, and 20. The activity was found to be about 10^{-4} _c/cm^3 which is not an unexpectedly high value.*[13]

[10] AI - 1959
[11] AI – 1961a
[12] AI - 1959
[13] AI - 1959

- *At 1530* [on July 12, 1959 during run 14], *both reactor room (high bay area) air monitors showed a sharp increase in activity. In an attempt to reduce the activity level, the reactor pressure was lowered to less than 1 psig from its former pressure of 2 psig. A survey of the loading face shield revealed that an excessive radiation reading existed over the reactor sodium level coil thimble located in core channel 7. The initial reading was 500 mr/hr. A high bay air sample had an activity of 3 x 10^{-7} _c/cm^3 after 15 min of decay and 4.5 x 10^{-8} _c/cm^3 after 90 min of decay. At 1620, it was noted that the filter from the air sampler showed an activity level of 160,000 c/m. At 1700, a sharp increase in the stack activity to 1.5 x 10^{-4} _c/cm^3 was noted. This returned to normal by 2200. By 1700, the radiation level over core channel 7 had reached 25 r/hr. It was decided to shut the reactor down and replace the thimble with a standard plug. Accordingly, at 1730, reduction of reactor power was begun. At 2057, the reactor was shut down, the drive units removed, and the cask placed in operation.*[14]

- *At 0900, on July 14, high bay activity increased to 14,000 cpm on the air monitor.*[15]

- *It seems quite likely that the first cladding failure occurred during the afternoon of the first day (July 12) of run 14. This conclusion is drawn from the observation that the radioactivity in the high-bay area (over the reactor loading face) increased markedly at this time and was almost certainly due to leakage of reactor cover gas into the area. It was believed that the time that a seal had failed on the sodium level probe. It now seems more likely that the leak had existed for some time and was suddenly noticeable because of the large increase in the radioactivity of the cover gas. The activity in the radioactive gas decay tanks also showed a sharp increase with the first samples taken after the start of run 14 on July 15 …. The activity in these tanks decayed continuously after July 15, indicating that most of the cladding failures had occurred by this time.*[16]

- *The first helium cover gas sample during run 14 which gave an indication of unusually high radioactivity was taken on July 18. Since the radiation level at the surface of the 2-liter sample chamber was ~30 mr/hr, no further attempt was made to assay the concentration quantitatively. … It is difficult to interpret cover gas samples subsequent to the July 26 shutdown since bleeding and flushing operations to the gas decay tanks and out the stack were almost immediately commenced.*[17]

- *At 0645 [on July 21, 1959], radioactivity in the reactor began building up, as indicated on the continuous air monitors. This buildup continued until at 1000 the two air monitors were reading 15,000 c/m and 18,000 c/m. At 1400, the results of a high bay air sample showed that the high bay activity level was 2 x 10^{-9} _c/cm^3.*[18]

- *On August 2, a parted fuel element from core channel 12 became lodged in the fuel handling cask. Attempts to dislodge it were not successful, so it was necessary to suspend the operations of examining the fuel. … A considerable amount of radiation exposure and*

[14] AI - 1959
[15] AI - 1959
[16] AI - 1959
[17] AI - 1962
[18] AI - 1959

contamination in the high bay area was resulting from the attempts to dislodge the jammed fuel element.[19]

- *The activity in the main primary gallery atmosphere, prior to run 14 was ~1 x 10^{-5} _c/cc. On August 11, 1959, following termination of run 14, a value of about 2 x 10^{-5} _c/cc was measured.*[20]

The post-event inquiries also reported some general findings about the radiological releases:

- *Considering a total accumulated burnup for the entire core of 2250 Mwd, assuming that 1 Mw corresponds to a fission of 1 gm of material per day, and using the calculated average release fraction of about 4 x 10^{-4}, it is calculated that a total of 0.9 gm of fission products were released to the sodium. This corresponds to a concentration in sodium of only 0.04 ppm.*[21]

- *No iodine was ever detected in reactor core gas samples.*[22]

- *Strontium-90 has deposited to the extent of about 0.8 _c/cm^2 in the primary system.*[23]

- *With the exception of the inert gases Xe^{133} and Kr^{85}, all of the fission fragments remained in the sodium, or were absorbed by the carbon or by the sodium-wetted metal surfaces.*[24]

- *As a result of the SRE fuel damage, reactor core gas activity was initially 10^{-3} _c/cc, but after extensive purging plus normal decay, activity stabilized around 10^{-6} _c/cc. … Since these values greatly exceeded tolerance levels, only a small amount of reactor cover gas could be allowed to pass into working areas.*[25]

- *The cold trap located in the primary system was effective in removing fission product contamination.*[26]

- *One would expect the release fraction of iodine to be considerably higher than that of cesium rather than one-third of it as actually found. … A possible answer to the low iodine value would be the escape of this element from the primary sodium to the cover gas system. However, iodine has never been found in the cover gas of the SRE either after this incident or in any of the many other gas samples taken during the operation of the reactor.*[27]

- *Although 5,000 to 10,000 curies of fission product activity were unexpectedly released to the primary sodium system no radiological emergency of any nature occurred.*[28]

---

[19] AI - 1959
[20] AI - 1959
[21] AI - 1959
[22] AI - 1959
[23] AI – 1961a
[24] AI – 1961a
[25] AI – 1961c
[26] AI - 1962
[27] AI - 1962
[28] AI - 1962

For radioactive material generated within the core of a nuclear reactor to harm workers and/or members of the public, the integrity of the tubes holding the nuclear fuel must be compromised allowing radioactivity to be released into the primary coolant, and then that radioactivity must somehow get out of the primary coolant into the environment. That the first condition was met is evident in the account of 5,000 to 10,000 curies of fission product activity being released into the primary sodium coolant.[29] The reports about high radiation levels in the SRE high bay provide compelling evidence that radioactivity escaped from the primary sodium coolant into the helium cover gas above the reactor pool and into the SRE building.

What were the potential pathways for radioactivity released from the damaged fuel elements and then the primary sodium coolant to reach the environment? Leading candidates were the ventilation system that handled the SRE building and the system used to vent gases from the reactor pool. The following section addresses these potential pathways.

SYSTEMS FOR PROCESSING AND RELEASING RADIOACTIVE GASES

SRE used helium gas to fill space above the liquid sodium in the reactor pool. The primary sodium fill-tank atmosphere was vented to the gaseous storage tanks. There were four gaseous storage tanks, each with a capacity of 100 psig of 2700 cubic feet. Helium gas flowed from the space above the reactor pool to the primary sodium fill-tank to the gaseous storage tanks primarily during three operating evolutions:

1. When the reactor was brought up to temperature causing the sodium level in the pool to rise, forcing some of the helium cover gas to the fill-tank and then the storage tanks.
2. Whenever the operating pressure is reduced.
3. Whenever the reactor cover gas is purged.[30]

Workers could discharge the contents of the gaseous storage tanks through a flow-rate meter and filters and out the building vent stack. The flow from the gaseous storage tank was diluted with about 25,000 cubic feet per minute of outside air before exhausting out the building vent stack. Procedures allowed discharges only when calculated levels at the stack exhaust would be less than 1×10^{-7} _c/cm^3. If the activity could be attributed solely to xenon, a release level of 1×10^{-5} _c/cm^3 is permitted.[31]

Radioactivity clearly reached the gaseous storage tanks following the fuel damage during run 14:

> *It seems quite likely that the first cladding failure occurred during the afternoon of the first day (July 12) of run 14. This conclusion is drawn from the observation that the radioactivity in the high-bay area (over the reactor loading face) increased markedly at this time and was almost certainly due to leakage of reactor cover gas into the area. It was believed that the time that a seal had failed on the sodium level probe. It now seems more likely that the leak had existed for some time and was suddenly noticeable because*

[29] This statement does not represent or imply endorsement of this range as the true amount of fission products released into the primary coolant. It merely indicates agreement that a release occurred – not to the magnitude of that release.
[30] AI - 1959
[31] AI - 1959

> *of the large increase in the radioactivity of the cover gas. The activity in the radioactive gas decay tanks also showed a sharp increase with the first samples taken after the start of run 14 on July 15 …. The activity in these tanks decayed continuously after July 15, indicating that most of the cladding failures had occurred by this time.*
>
> and
>
> *It was decided to pressurize and vent the reactor atmosphere once in order to reduce the radioactivity level caused by the xenon in the cover gas. At 0550, July 15, the reactor pressure was reduced from 1.8 psig to 0.6 psig, repressurized to 3.0 psig, and then reduced to 1 psig.*
>
> and
>
> *The first storage tank sample taken on July 15, 1959, after the start of run 14, indicated an extremely high activity; so high in fact that the counter had not been calibrated in that range.*[32]

The gaseous storage tank sample taken on July 15th was so radioactive that it exceeded the calibration range of the instrument used to process the sample. A similar problem was encountered three days later when the first sample was taken of the helium cover gas above the reactor pool:

> *The first helium cover gas sample during run 14 which gave an indication of unusually high radioactivity was taken on July 18. Since the radiation level at the surface of the 2-liter sample chamber was ~30 mr/hr, no further attempt was made to assay the concentration quantitatively.*[33]

The helium cover gas sample taken on July 18th was so radioactive that it precluded further analysis.

High levels of radioactivity released from the damaged fuel on July 13th into the primary sodium reached the cover gas and the gaseous storage tanks. Unfortunately, no data was found in the documents reviewed regarding the number of (or absence of) discharges from the gaseous storage tanks following the July 13th event. Thus, it is impossible to confirm or refute the assertion that "no radiological hazard was present to the reactor environs" via the gaseous storage tank pathway.

Turning to the potential pathway from the SRE building itself; namely, the high bay area. The SRE structure around the reactor was not one of the steel-lined, reinforced concrete containments used in commercial nuclear power reactors. Instead:

> [It] *was a low-air-leakage building with ventilation and exhaust systems to control leakage and air flow paths. The interior of the building was maintained at a lower pressure than the exterior of the building so that air flow would always be into the building. In this manner, any radioactive particulates that might have escaped would have been retained within the structure and trapped by the exhaust filters.*
>
> and

[32] AI - 1959
[33] AI - 1962

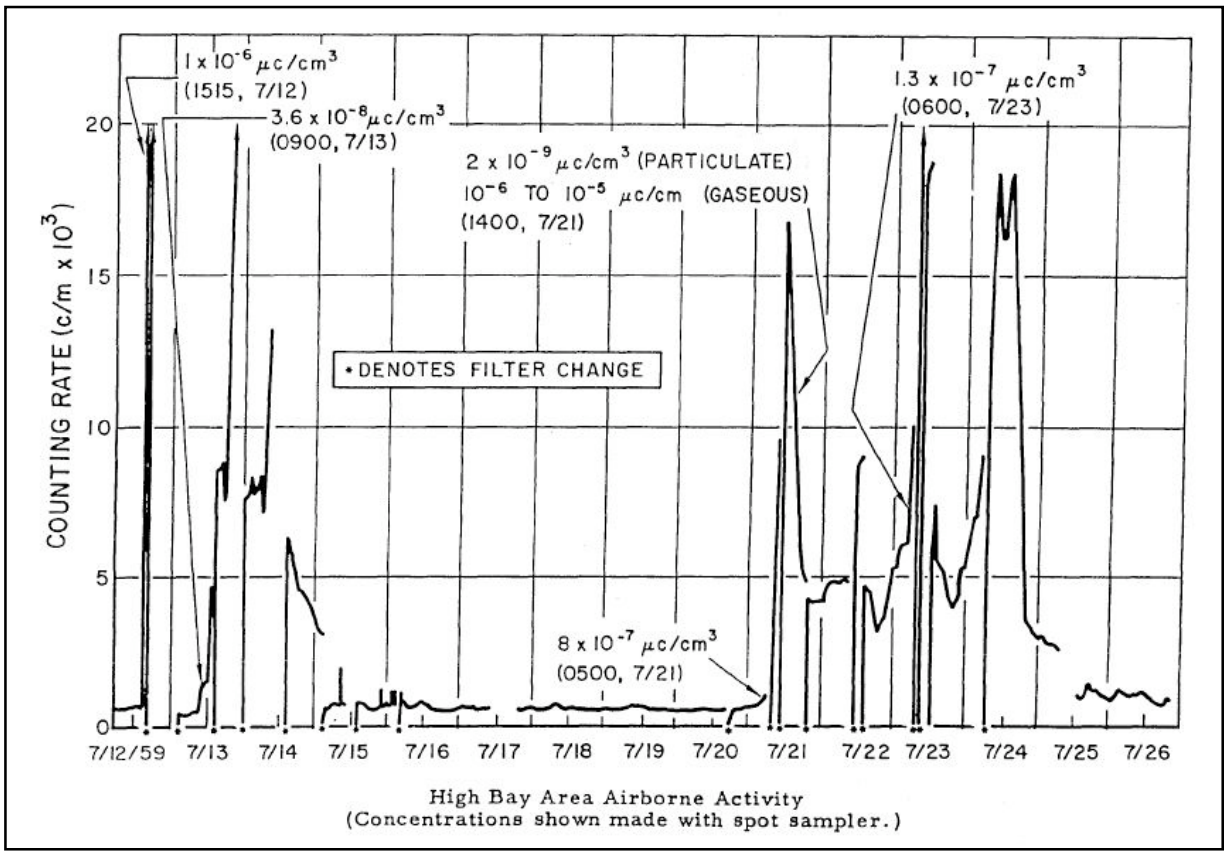
The building ventilation system was designed and operated so that air moved toward potentially contaminated areas. Makeup air was brought in from the outside and combined with recirculated air within the administrative areas to maintain positive pressure relative to the contaminated areas. Fresh air at the rate of five air changes per hour was supplied to the reactor room by independent supply fans. Exhaust fans and high-efficiency filters on the reactor room roof were sized to maintain the reactor room pressure below all contiguous regions, which were (1) administration areas, (2) hot cell, and (e) out of doors. The issue of filters reduced the possibility of local contamination by the accumulation of radioactive particulates on building and equipment surfaces.[34]

Data show that radioactivity reached the high bay area following the July 13th power excursion:

At 0900, on July 14, high bay activity increased to 14,000 cpm on the air monitor.

and

At 0645 [on July 21, 1959], *radioactivity in the reactor began building up, as indicated on the continuous air monitors. This buildup continued until at 1000 the two air monitors were reading 15,000 c/m and 18,000 c/m. At 1400, the results of a high bay air sample showed that the high bay activity level was 2 x 10^{-9} _c/cm^{3}."*[35]



[34] Rockwell - 1983
[35] AI - 1959

ACTIVITY HISTORY OF THE REACTOR COVER GAS

Date	Specific Activity ($\mu c/cm^3$)	
December 11, 1958	1.9×10^{-3}	
December 14, 1958	3.0×10^{-4}	
December 15, 1958	1.0×10^{-3}	
February 14, 1959	1.7×10^{-3}	
February 26, 1959	0.06	(taken shortly after end of run)
May 26, 1959	1.7×10^{-3}	
June 13, 1959	2.4×10^{-4}	
June 16, 1959	1.8×10^{-5}	
June 20, 1959	3.8×10^{-5}	(during N_2 stripping operation)
July 18, 1959	-	(sample chamber read 30 mr/hr at surface pressure)
July 29, 1959	-	(reactor cover gas bled down to negative)
August 1, 1959	5.5	
August 12, 1959	0.87	
August 19, 1959 August 20, 1959	-	(cover gas purged on August 19 and 20)
August 25, 1959	0.05	
August 28, 1959	0.082	(this sample showed no decay in 48 hr)
August 30, 1959	0.082	
September 1, 1959	0.085	
September 16, 1959	0.07	(cover-gas purging operation started)
September 22, 1959	0.01	
September 24, 1959	0.01	
September 29, 1959	4.0×10^{-3}	

RADIOACTIVE CONCENTRATIONS IN GAS DECAY TANKS

Date	Activity ($\mu c/cm^3$)
November 18, 1958	$< 10^{-7}$
November 19, 1958	$< 10^{-7}$
November 23, 1958	$< 10^{-7}$
November 24, 1958	$< 10^{-7}$
November 25, 1958	5×10^{-7}
November 26, 1958	$< 10^{-7}$
November 27, 1958	$< 10^{-7}$
December 2, 1958	4.6×10^{-7}
December 3, 1958	4.6×10^{-7}
December 9, 1958	6×10^{-6} (washed fuel from R-9)
December 10, 1958	5×10^{-6}
December 11, 1958	8×10^{-6}
December 12, 1958	1.2×10^{-4} (washed fuel from R-10)
December 13, 1958	2.4×10^{-4}
December 14, 1958	3.0×10^{-5}
December 15, 1958	4.4×10^{-6}
December 16, 1958	2×10^{-5}
December 17, 1958	2×10^{-6}
December 18, 1958	6×10^{-6}
December 19, 1958	7×10^{-3} (first measurement after restart of run 8)
December 20, 1958	7×10^{-5}
December 21, 1958	1.5×10^{-5}
December 22, 1958	1.4×10^{-5}
December 23, 1958	4×10^{-6}
December 24, 1958	4×10^{-5}
December 26, 1958	8×10^{-5}
December 27, 1958	2×10^{-5}
December 28, 1958	8×10^{-6}
December 29, 1958	2×10^{-6}
December 30, 1958	2.7×10^{-6}
July 25, 1959	0.08
July 27, 1959	0.04
July 28 - Aug. 22	bleeding at 4 cfh
August 22, 1959	0.07
Aug. 22 - Sept. 16	bleeding at 4 cfh
September 16, 1959	0.065

As with the above case of the potential pathway through the gaseous storage tanks, the data do not reveal what the radioactivity levels in the effluent from the building exhaust fans. Once again, it is impossible to confirm or refute the assertion that "no radiological hazard was present to the reactor environs" via the SRE building ventilation exhaust pathway.

The scant, disconnected data prevents a quantitative assessment of the radioactivity released to the atmosphere following the extensive fuel damage experienced during SRE run 14. The data show that large amounts of radioactivity released from the damaged fuel elements reached (a) the helium cover gas above the reactor pool, (b) the high bay area, and (c) the gaseous storage tanks.

That large amounts of radioactivity reached the helium cover gas above the reactor pool is evident from the table titled Activity History of the Reactor Cover Gas. The radioactivity level increased by a factor of approximately one million (1,000,000) between the samples taken on June 20, 1959, and August 1, 1959. The actual increase was almost certainly much greater because (a) the reactor ceased operation on July 26, 1959, providing nearly five (5) days for decay of short-lived radioisotopes before the August 1st sample, and (b) repeated vent and purge operations were performed on the helium cover gas in mid-July 1959 following the July 13th transient, permitting transfer of considerable radioactivity to the gaseous storage tanks before the August 1st sample. The radioactivity in the helium cover gas could leak into the high bay area or be vented to the gaseous storage tanks.

That large amounts of radioactivity reached the high bay area is illustrated in the figure titled High Bay Area Airborne Activity. The counting rate of approximately 1,000 counts per minute on July 12th soared nearly 20-fold during the July 13th transient, fell back down towards 1,000 counts per minute during the helium cover gas venting evolutions, and re-soared nearly 20-fold later in July when the reactor was restarted.

That large amounts of radioactivity reached the gaseous storage tanks is evident from the table titled Radioactive Concentrations in Gas Decay Tanks. The radioactivity levels increased by nearly a factor of 30,000 following the fuel damage incurred on July 13th.

The evidence of large amounts of radioactivity reaching the high bay area and the gaseous storage tanks clearly demonstrates that the second containment barrier – namely, the primary coolant loop – between fission products produced by the nuclear fuel during reactor operation and the environment had been breached after 13 of the 43 fuel elements failed during run 14

The third and final containment barrier was the ventilation system that processed the high bay area atmosphere and the contents of the gaseous storage tanks through filters prior to discharge via the building stack. No data was found on either the radiation levels at the stack release point or on the number, timing, and radioactivity levels of releases from the gaseous storage tanks. The only information – albeit indirect – covered the radiation levels inside the ventilation system ductwork.[36] This data, from 1966, indicated the radiation levels measured in ductwork upstream of filters was 2 to 20 times the radiation levels measured downstream of the filters. The data clearly demonstrate (a) the ventilation system filters were effective in removing radioactivity from the process flows, and (b) the ventilation system filters did not remove all radioactivity from the process flows.

[36] Rockwell – 1983, Table 7

ACTIVITY LEVELS OF SRE VENTILATION SYSTEM (1966)

Sample	Description and Location	β-γ Activity (dpm/100 cm^2)
1	West duct, upstream of filter	2,334
2	West duct, downstream of filter	129
3	Center duct, upstream of filter	10,181
4	Center duct, downstream of filter	1,293
5	East duct, upstream of filter	756
6	East duct, downstream of filter	423
7	East plenum floor, under filter	1,953
8	Center plenum, under filter	1,479
9	West plenum floor, under filter	2,118

The data, however, are inconclusive with regard to the key question of whether the ventilation system served as an effective containment barrier against the release of radioactivity to the atmosphere in harmful amounts. The data do not permit a quantitative analysis and prompt a turn to a qualitative assessment.

The qualitative assessment was performed using information from a subsequent fuel damage event at another sodium-cooled, graphite moderated nuclear reactor called the Enrico Fermi Atomic Power Station (or Fermi-1) in 1966 and from a literature search for experimental work/analyses.

There were many similarities between SRE and Fermi. In both cases, the Xenon-133 and Krypton-85 isotopes were found in the cover gases, but no radioiodine was detected. The amount of fission products released into the primary sodium at SRE has been estimated at 5,000 to 10,000 curies, corresponding closely to the 10,000 curies estimated to reach the primary sodium from the damaged fuel at Fermi.[37]

There are also many differences between SRE and Fermi. The radiation levels from the primary loop piping were 3 rem/hour at SRE and 3 millirem/hour at Fermi. The radiation levels from the intermediate heat exchanger (between the primary and secondary loops) were 11 rem/hour at SRE and 6 millirem/hour at Fermi. And the radiation levels from the primary cold trap were 81 rem/hour at SRE and 40 millirem/hour at Fermi.[38]

[37] AI - 1969
[38] AI - 1969

If the amount of fission products released into the primary sodium was approximately 10,000 curies at both SRE and Fermi, why were the radiation levels at primary loop components so disparate? Some theorize that the fact that the SRE sodium was corrupted with tetralin whereas the Fermi sodium was "pure" accounted for the difference. In this theory, the tetralin and its byproducts acted as a getter to capture the fission products and cause them to plate out on component surfaces at a greater rate.[39]

Another explanation might be that the amount of fission products released into the SRE primary sodium was significantly higher than amount released into the Fermi sodium. Instead of the tetralin and its byproducts removing more fission products from the SRE sodium, it might have been there were far more fission products released into the SRE sodium. Support for this explanation can be found:

> *The Na^{24}* [Sodium-24, a radioactive isotope with a 15-hour half-life] *activity in the primary coolant delayed the procurement of the first sodium sample until August 2, about 7 days after reactor shutdown. It should be noted that during this interval, circulation and cold-trapping of the primary sodium continued; thus the loss of some portion of the fission product activity originally present prior to obtaining the first sample was certainly possible.*[40]

Much of the SRE radioactive release inventories were based upon the activity levels found in this August 2nd sodium sample – but this sample (a) lacked the fission products that escaped to the helium cover gas and were removed by the venting/purging evolutions performed in mid-July after the power excursion and (b) lacked the fission products that were removed from the primary sodium via the cold trap or plate-out on piping and components. Thus, the actual amount of fission products released from the damaged fuel elements might have significantly exceeded the 5,000 to 10,000 curie range.

The reported inability to detect radioiodine in the cover gas at SRE and Fermi-I is unexplained and inconsistent with numerous experimental findings. For example:

> *Two series of experiments have been conducted to obtain the gas-liquid equilibrium partition coefficient k sub d and the nonequilibrium partition coefficient k exp sub d of volatile fission products such as Ce, iodine, and Te between liquid Na and the gas phase. … Cesium shows the largest k sub d (20-100). The k sub d value of I scatters as widely as 0.02-0.5 at 450 deg C and 0.3-0.8 at 650 deg C.*[41]

> *Among the fission products released in a large-scale fuel failure incident, the iodine isotopes, together with rare gases, will rise through the sodium pool in bubbles. Some of the iodine will react with sodium vapor present in the bubbles, and with liquid sodium at the bubble boundary, to form sodium iodide (NaI), and dissolve into the sodium pool. The rest of the iodine will remain in the bubble with the rare gases, and without being trapped in sodium, will escape from the pool surface and directly reach the cover gas.*

[39] AI - 1969
[40] AI - 1962
[41] Haga - 1992

Iodine retained in the sodium in the sodium pool in the form of NaI will also ultimately tend to escape from the pool surface to the cover gas.[42]

Irrespective of defect size, noble gases (Xe, Kr) are released fairly quickly and their limited solubility in sodium ensures rapid removal to gas space. Fission products of highly volatility behave in a similar manner and those elements of high yielding, namely Cs, I, Ba and Sr can also be released during the initial stages of fuel clad failures.[43]

The level of activity circulating in a coolant depends upon the solubility behavior of various isotopes. In principle, levels of activity produced by partly soluble Te, Sb, Sn, Ag radionuclines and I can be reduced by operations of cold trap. However, if fuel pin failures become excessive, there is possibility that cold trap may become a relatively ineffective sink and plate-out of various products may occur elsewhere in a circuit.[44]

Cold trap is used as purification method for sodium in the first loop of LMFBRs as well as for potassium in the second loop. Although the principal role of cold trap in LMFBRs is to control the concentration of dissolved oxygen impurity and hence maintain acceptable levels of radioactive corrosion products, a number of traps have been known to collect radioactive impurities. Decreasing of sodium temperature in cold trap causes crystallization of Na_2O on the wires of steel mesh.[45]

KALIMER Source terms: Noble gas (Xe, Kr) 100%, Halogens (I, Br) 10%, Alkali metals (Cs Rb) 10%, Te group (Te, Se, Sb) 10%, Noble metals (Ru, Rh, Mo, Tc) 1%, Ba and Sr 1% [46]

The KALIMER experiment (2003) was conducted with 100 percent of the noble gases (xenon and krypton), 10 percent of the halogens (iodine and bromine), 10 percent of the alkali metals (cesium), 1 percent of the noble metals (ruthenium), and 1 percent of the strontium released from damaged fuel into the primary sodium. The work by Hasan (also 2003) indicated that virtually all of the non-soluble noble gases released into the primary sodium reached the cover gas while other fission products reached the cover gas depending on their solubility rates in sodium. The work by Haga (1992) indicated that the sodium to cover gas transfer rate for radioiodine was strongly temperature dependent. The temperature-dependence may have played a role during the helium cover gas purging/venting conducted at the SRE in mid-July following the July 13th power excursion.

CONCLUSIONS

The first containment barrier between fission products and the environment – the fuel cladding – was breached when 13 of 43 fuel elements in the SRE reactor core overheated and failed during run 14. Fission products were released into the primary sodium.

---

[42] Tashiro - 2001
[43] Hasan - 2003
[44] Hasan - 2003
[45] Hasan - 2003
[46] Cho - 2003

The second containment barrier – the primary sodium – was compromised when radioactivity escaped into the high bay area and flowed to the gaseous storage tanks.

The third and final containment barrier – the ventilation system – was compromised because some radioactivity reached the atmosphere from the high bay area and from the gaseous storage tanks.

Scant and disconnected data prevented a quantitative assessment of what got out when. Instead, the maximum and minimum radioactive release fractions will be determined.

The upper bound on the amount of gaseous radioactivity that reached the environment is established by the percentage of fuel elements damaged during run 14 at SRE. If all the gaseous radioactivity from the 13 damaged fuel elements reached the environment, then 30 percent of the SRE core inventory was released (based on the entire contents from 13 of 43 fuel elements).

The lower bound on the amount of gaseous radioactivity that reached the environment is established by taking the upper bound and accounting for (a) the likelihood that not all of the gaseous radioactivity was released from the fuel elements into the primary sodium and (b) the likelihood that not all of the gaseous radioactivity eluded the hold-up and filter containment of the ventilation system to reach the atmosphere. Experimental results[47] suggest a release fraction of 10 percent is reasonable. The data from decommissioning the ventilation system ductwork in 1966[48] suggest a release fraction of 10 percent is also reasonable, at least for cesium (Cs-137). The lack of charcoal absorber units in the filter train of the ventilation system means that essentially none of the iodine (I-131) would be removed from the effluent stream. The lower bound is therefore:

$Cs\text{-}137_{LOWER\ BOUND}$ = Upper bound x Release fraction x Ventilation system attenuation
= 30 percent x 0.10 x 0.10 = 0.3 percent

$I\text{-}131_{LOWER\ BOUND}$ = Upper bound x Release fraction x Ventilation system attenuation
= 30 percent x 0.10 x 1.00 = 3 percent

The range of releases to the environment following the fuel damage during run 14 is 0.3 to 30 percent for Cs-137 and 3 to 30 percent for I-131.

It is almost certain that all the gaseous radioactivity did not escape from the damaged fuel elements to the primary sodium, from the primary sodium to the cover gas, and from the cover gas to the atmosphere. It is also unlikely that the releases were minimized all the way to the lower bound. The actual releases are somewhere between these end points.

Factors driving the answer towards the upper bound include (a) the fact that the evidence strongly suggests the primary sodium boiled, thus providing a mechanism for radioiodine and other gases to avoid retention by the liquid sodium and escape directly to the helium cover gas inside the bubbles, (b) the helium cover gas was vented/purged to the gaseous storage tanks soon after the onset of significant fuel damage, (c) the fact that unlike virtually all other instances

[47] Cho - 2003
[48] Rockwell – 1983, Table 7

involving significant amounts of fuel damage, the SRE reactor core continued to be operated for many days, and (d) the fact that unusually high radioactivity levels were present in the high bay area and in the stack on July 12th and 13th. Factors driving the answer towards the lower bound include (a) the ability of liquid sodium to bond with radioiodine to form NaI, (b) the subsequently shown propensity for tetralin to act as a getter and absorb fission products from the liquid sodium, and (c) the reported decay of radioactivity measured in the gaseous storage tanks after July 15th.

These factors tend to balance one another. Consequently, it seems reasonable that assuming a value near the midpoint – say 15 percent – is closer to the actual release fraction than either the upper or lower bound. It is both unsatisfying and frustrating not to be able to develop a more definitive basis for the estimated release fraction, but the scant and disconnected data prevent such analysis.

Assessment performed by:

David A. Lochbaum

DOCUMENTS REVIEWED AND CITED

R. L. McKisson and K. E. Horton, Atomics International, "The Behavior of Tetralin in Liquid Sodium," NAA-SR-1771, February 1, 1957.

C. Rogers McCullough, Chairman – Advisory Committee on Reactor Safeguards, to John A. McCone, Chairman – Atomic Energy Commission, "Comparison of Safety Features of GE-Vallecitos Boiling Water Reactor (VBWR) / Sodium Graphite Reactor Experiment (SRE) / Shippingport Pressurized Water Reactor (PWR) / Rural Cooperative Power Association Reactor, Elk River, Minnesota," December 15, 1958. Cited as "ACRS – 1958"

A. A. Jarrett, General Editor, Atomics International, "SRE Fuel Element Damage: An Interim Report," NAA-SR-4488, November 30, 1959. Cited as "AI – 1959"

R. L. Ashley, R. J. Beeley, F. L. Fillmore, W. J. Hallett, B. R. Haywood Jr., and A. A. Jarrett, Atomics International, "SRE Fuel Element Damage: Final Report," NAA-SR-4488 (suppl), June 30, 1961. Cited as "AI – 1961a"

F. L. Fillmore, Atomics International, "Analysis of SRE Power Excursion of July 13, 1959," NAA-SR-5898, September 15, 1961. Cited as "AI – 1961b"

J. L. Ballif, Atomics International, "Metallurgical Aspects of SRE Fuel Element Damage Episode," NAA-SR-4515, October 15, 1961.

W. J. Freede, Atomics International, "SRE Core Recovery Program," NAA-SR-6359, December 1, 1961. Cited as "AI – 1961c"

R. S. Hart, Atomics International, “Distribution of Fission Product Contamination in the SRE,” NAA-SR-6890, March 1, 1962. Cited as “AI – 1962”

G. B. Zwetzig, Atomics International, “Survey of Fission and Corrosion Product Activity in Sodium or NaK Cooled Reactors,” AI-AEC-MEMO-12790, February 28, 1969. Cited as “AI – 1969”

J. W. Carroll, C. C. Conners, J. M. Harris, J. M. Marzec, and B. F. Ureda, Rockwell International, “Sodium Reactor Experiment Decommissioning Final Report,” ESG-DOE-13403, August 15, 1983. Cited as “Rockwell – 1983”

K. Haga, Y. Nishizawa, T. Watanabe, S. Miyahara, and Y. Himeno, “Equilibrium and Nonequilibrium Partition Coefficients of Volatile Fission Products Between Liquid Sodium and the Gas Phase,” Nuclear Technology, Vol. 97, No. 2, pp. 177-185, February 1992. Cited as “Haga – 1992”

Suguru Tashiro and Norihiko, Ibaraki University, “Diffusion Coefficient Determination of Sodium Iodide Vapor in Rare Gases with Use of Ionization Sensor,” Journal of Nuclear Science and Technology, Vol. 38, No. 7, pp. 551-556, July 2001. Cited as “Tashiro – 2001”

Ahmed Hasan, Yasser T. Mohamed, and Tarek F. Mohammaden, New Mexico Technological University, “Waste Generated from LMR-AMTEC Reactor Concept, Waste Management Conference, Tuscon Arizona, February 23, 2003. Cited as “Hasan – 2003”

Young Ho Cho, Mee Jang, Young Wook Lee, Moon Soo Park, Chang Sun Kang, Chang Hyun Chung, Do Hee Hahn, and Joo Hyun Moon, “The Analysis of Radiological Consequences Due to Hypothetical Core Disruptive Accident in KALIMER,” Paper 3074, Proceedings of ICAPP ’03, May 2003. Cited as “Cho – 2003”