

EMERGENCY CORE COOLING SYSTEM SUMP CHEMICAL EFFECTS ON STRAINER HEAD LOSS

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ABSTRACT

Chemical precipitates formed in the recovery water following a Loss of Coolant Accident (LOCA) have the potential to increase head loss across the Emergency Core Cooling System (ECCS) strainer, and could lead to cavitation of the ECCS pumps, pump failure and loss of core cooling. Atomic Energy of Canada Limited (AECL) has been involved in the investigation of chemical effects on head loss for its CANDU[®] and PWR (Pressurized Water Reactor) customers.

The chemical constituents of the recovery sump water depend on the combination of chemistry control additives, fission products, radiolysis products (e.g., nitric acid), and the corrosion and dissolution products from metals, concrete, and insulation materials. Some of these dissolution and corrosion products (e.g., aluminum and calcium) may form significant quantities of precipitates. The presence of chemistry control additives such as lithium and sodium hydroxide, trisodium phosphate (TSP) and boric acid can significantly influence the precipitates formed. While a number of compounds may be shown to be thermodynamically possible under the conditions assumed for precipitation, kinetic factors play a large role in the type and morphology of precipitates observed. Precipitation is also influenced by insulation debris, which can trap precipitates and act as nucleation sites for heterogeneous precipitation.

This paper outlines the AECL approach to resolving the issue of chemical effects on ECCS strainer head loss, which includes modeling, bench top testing and reduced-scale testing; the latter conducted using a temperature-controlled variable-flow closed-loop test rig that includes an AECL *Finned Strainer*[®] test section equipped with a differential pressure transmitter. Models of corrosion product release and the types of precipitates expected in post-LOCA sumps are discussed. Finally, this paper discusses reduced-scale test results and presents a possible method for chemical effects head loss modeling.

1. BACKGROUND ON THE ECCS STRAINER CLOGGING ISSUE

In the event of a LOCA, emergency shutdown systems activate and very quickly stop the neutron chain reaction. Heat removal, normally performed by the reactor coolant that is now spilling into containment, is handled by the ECCS, a system designed to remove decay heat and cool the core. In CANDU plants, short-term cooling is provided by high- and medium-pressure injection systems with their own supplies of water [1]. The dousing spray system may also activate, controlling containment pressure by condensing steam with large volumes of cool water. Long-term cooling is provided by the low-pressure injection system, which uses water from the containment recovery sump to cool the reactor. The water in the recovery sump has many

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sources, including reactor coolant, ECCS tank water and dousing tank water. On the initial break, the reactor coolant flashes from high pressure to containment pressure, producing a two-phase steam and water jet. This jet could dislodge significant quantities of fibrous insulation that could challenge the operation of existing ECCS strainers.

The function of the ECCS strainer is to filter solids, typically insulation debris dislodged by the jet issuing from the line break, but also dirt, paint chips, signs and other debris, from the recovery water. After passing through the strainers, the water is pumped through heat exchangers and back into the core to provide long-term cooling. Without the strainers, debris would quickly clog the pumps and heat exchangers. Thus, the strainers provide a passive but important role in the long-term reactor core cooling function provided by the ECCS.

Depending on the quantity of debris and the size of the strainers, the strainers could become clogged, resulting in a large pressure drop across the strainer that could then cause structural deformation, insufficient pump suction head, pump cavitation and pump failure. In the 1990s, three nuclear plants experienced minor incidents that resulted in their ECCS becoming engaged but operation of the strainers was significantly hindered by debris [2]. Many countries, including Canada, began programs to address deficiencies in the ECCS strainer knowledge base [1], which eventually saw utilities replace strainers with larger, oversized units by 2003 [3].

Around the time that new installations were nearing completion, the scientific community began to identify chemical precipitates as another form of debris not yet considered. In an assessment performed by Los Alamos National Laboratory (LANL) [4], it was noted that evaluations of hydrogen generation for Design Basis Accidents (DBA) include contributions from zinc and aluminum corrosion, although the effects of corrosion by-products were not yet considered for strainer performance. In a broad-scoped letter issued by the U.S. Nuclear Regulatory Commission (USNRC) [2] regarding ECCS performance, the regulator identified that chemical effects may have an effect on head loss and that addressees should consider these effects in their responses. Small-scale [5] and medium-scale tests [6] were conducted at LANL to investigate the issue. The Canadian Nuclear Safety Commission (CNSC) responded to the mounting evidence by raising GAI 06G01, "ECC Strainer Deposits".

AECL has been involved in the investigation of chemical effects on head loss for its CANDU and Pressurized Water Reactor (PWR) customers. Differences in containment chemistries have resulted in significantly different experiences when dealing with these different reactor types.

2. THE AECL APPROACH TO CHEMICAL EFFECTS

In order to evaluate the effects of sump water chemistry on strainer performance, AECL has developed the methodology shown in Figure 1. This methodology is similar in general philosophy but different in details of implementation from the WCAP (Westinghouse Commercial Atomic Power) method described by Lane et al. [7].

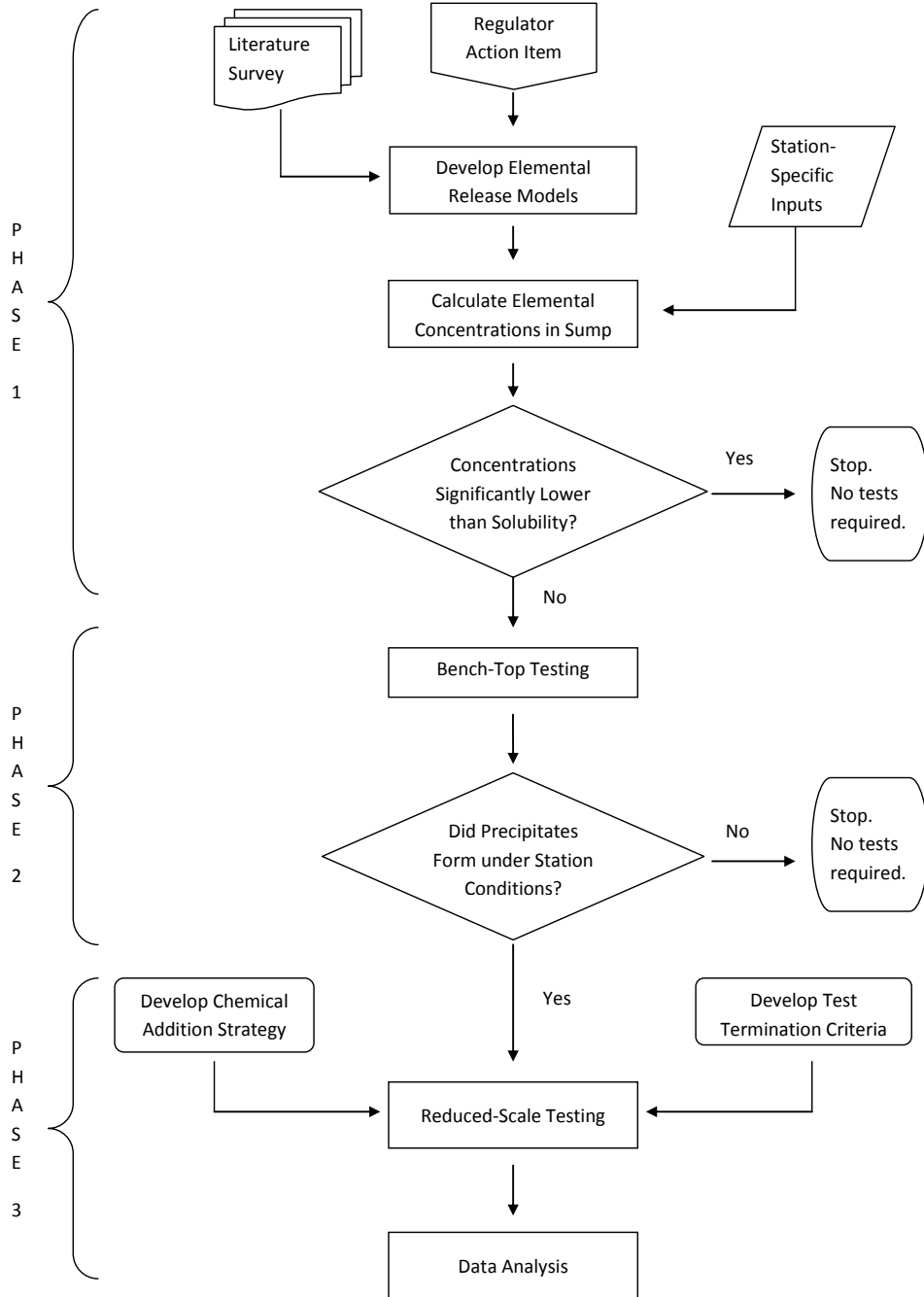


Figure 1. AECL’s General Strategy for Chemical Effects Resolution

Phase 1 begins the resolution of the regulatory action item (sump screen blockage) through a literature survey and development of release models in order to predict the concentrations of various chemical species in the sump. In the U.S., release models were developed by Lane et al. [7] for calcium, aluminum and silicate, as the predominant precipitates observed in the WCAP tests [7] were aluminum hydroxide (or aluminum oxyhydroxide)¹, sodium aluminum silicate and

¹ The observed precipitate may have been either aluminum hydroxide (Al(OH)₃) or aluminum oxyhydroxide (AlOOH); the SEM/EDS (Scanning Electron Microscope / Energy Dispersive X-ray Spectroscopy) data collected could not have provided conclusive evidence of the stoichiometry.

calcium phosphate. To create an aluminum release rate data set that covered most regions of interest, Lane et al. [7] conducted short-term corrosion tests on aluminum sheet in borated solutions at pH 4, 8 and 10 and compiled literature data from other longer-term aluminum corrosion tests to help fill in gaps. Despite significant scatter in the data (Figure 2), an empirical model (1) for aluminum release was produced that became the US industry standard. Using a very similar data set, AECL produced a semi-empirical equation (2) based on a first principles understanding of the overall kinetics. Since the AECL model requires aluminum release to behave in a consistent manner with respect to pH and temperature, outlying data (or data exaggerated due to short test duration) were not given as much weight. Therefore, depending on the conditions, the two models may differ, though both are within the scatter of the data. The differences between the models are mainly a result of the lack of agreement of the available data, likely caused by the differences in testing methods and test durations used by the separate groups, and the inherent repeatability of corrosion tests. AECL has observed experimental uncertainties of about 30% in nominally identical tests.

$$RR_{Al} = 10^{A-B/T+C \cdot pH^2 - D \cdot pH \cdot T} \quad (1)$$

$$RR_{Al} = A \times \exp(B \cdot pH) \times \exp(-C/T) \quad (2)$$

Where RR_{Al} is the release rate of Al, and A, B, C and D are constants.

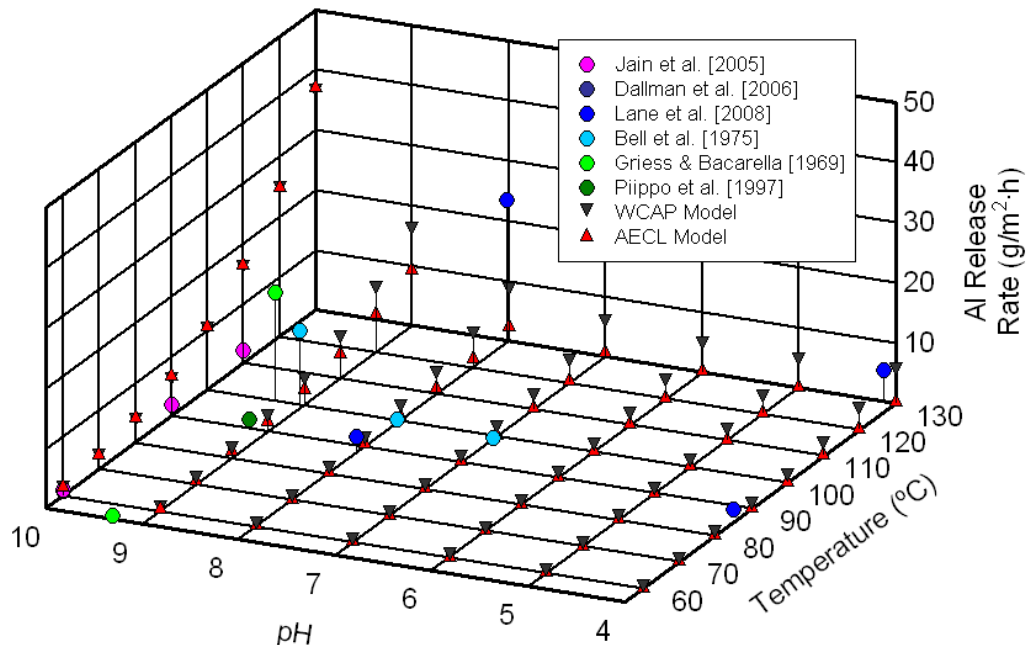


Figure 2. Data for aluminum corrosion (release) rate in borated solution and models developed by Westinghouse and AECL to describe the data.

Both models reasonably predict the aluminum concentration data reported by Dallman et al. [6] for Tests 1 and 5 of the Integrated Chemical Effects (ICE) Test Project (Figure 3). In these tests, large 12" square aluminum coupons were placed on racks, with 56 coupons sprayed by pH 11 NaOH solution or exposed to such spray for 4 hours and 3 coupons submerged for 30 days. In

Test 1 the submerged coupons were exposed to pH 9.5 borated water at 60°C, while in Test 5 the pH was 8-8.5. It can be shown by consideration of coupon weight change, mass balances and short spray duration that very little dissolved aluminum came from the sprayed coupons, making the lower curves in Figure 3 pertinent to the present analysis. It can be seen that the predicted 30-day release is in general agreement with the observed concentrations. To ensure conservatism in calculations of post-LOCA aluminum release for client utilities, maximum pH and temperature profiles are often used, and sprayed surfaces are assumed to contribute corrosion products to the sump for the duration of spray operation.

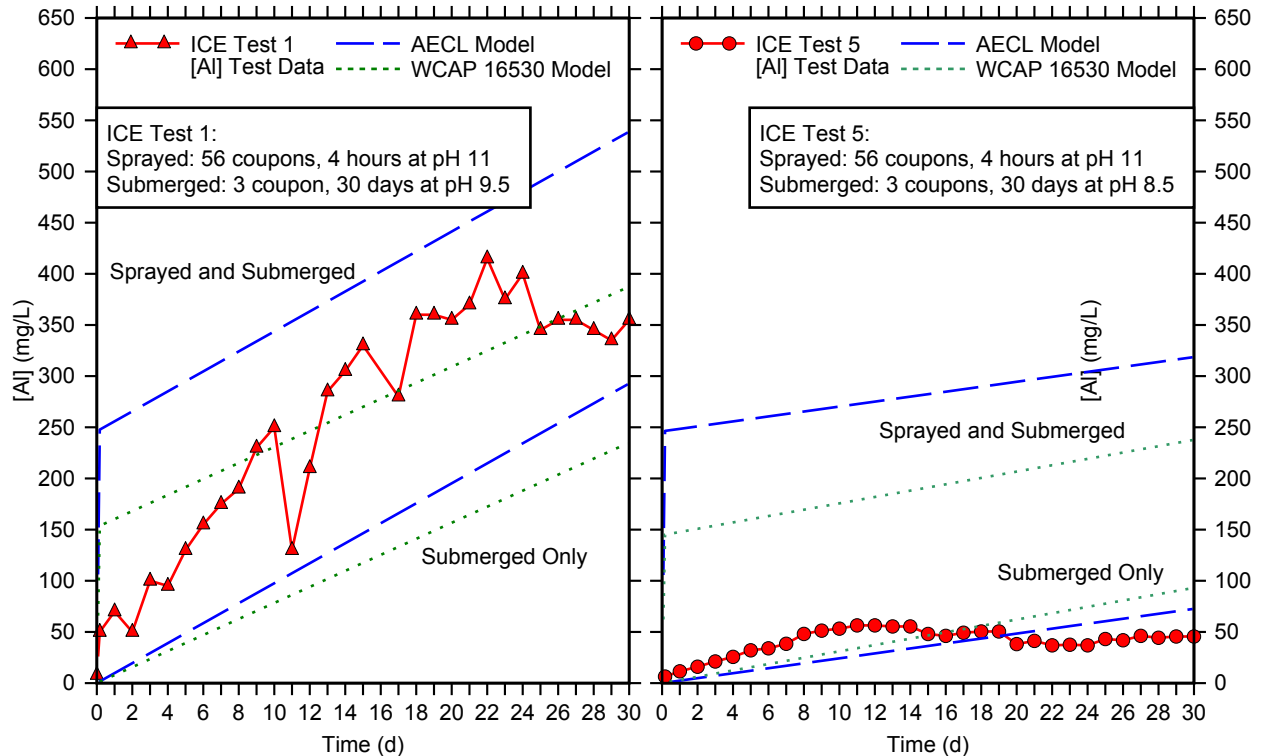


Figure 3. WCAP and AECL aluminum release models' predictions of ICE Test 1 (left) and Test 5 (right) aluminum concentration. ICE test concentration data adapted from Ref [6]. Spray pH, reported as < 12, was taken to be 11 for calculations. Upper curves are the calculated release for 3 submerged and 56 sprayed coupons, and lower curves are the calculated release for 3 submerged coupons only.

The aluminum release data developed for the U.S. nuclear industry in borated solutions could not be applied to the Canadian nuclear industry because aluminum release rates obtained in borated solutions are significantly higher than those reported for non-borated solutions. Therefore, AECL performed detailed aluminum release testing for the CANDU fleet under representative CANDU post-LOCA conditions. The 4- to 90-day tests examined the effects of pH, temperature, CO₂, hydrazine, cal-sil, TSP and alloy type on aluminum release rates from corroding coupons. The model developed was mainly a function of pH, temperature and time of the form shown in Equation (3). Note that the release rate has parabolic time dependence, which is a common feature of long-term corrosion tests resulting from the formation of an oxide film.

$$RR_{Al} = A \times f(T) \times \exp(B \cdot pH) \times t^{-0.5} \quad (3)$$

Once an appropriate release model has been developed, postulated post-LOCA temperature and pH profiles are used to determine release rates of the major precipitants, typically aluminum and calcium, as a function of time. Note that calcium in the sump water is not generally a concern except in the presence of phosphate. Release rates are converted to accumulated releases, and accumulated releases to sump concentrations. An important step in the AECL approach to chemical effects, and the last step of Phase 1, is to compare the calculated concentrations of precipitants to the solubilities of potential precipitates.

Precipitation will only occur when the concentrations of species in solution exceed the solubility limit with respect to a solid phase. This will not occur for some period after the start of the accident because of the kinetics of the various corrosion or dissolution reactions. Two scenarios are possible:

1. At constant temperature and pH, the concentrations of the relevant species increase in solution due to release from dissolution or corrosion until the solubility limit for the precipitating phase is exceeded (e.g., condition A in Figure 4), or
2. A change in temperature or concentration results in a decrease in the solubility of the precipitating phase such that it is now lower than the solution concentration (e.g., condition B in Figure 4).

Typically, some degree of supersaturation is required before precipitation occurs (Figure 4). Clearly in Scenario 2 a much higher degree of supersaturation can occur, increasing the likelihood of precipitation.

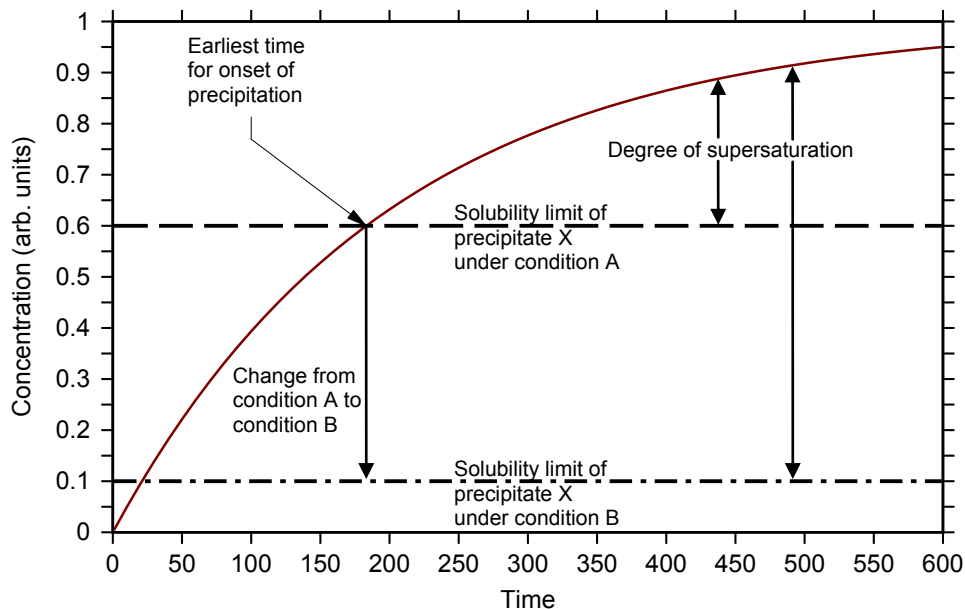


Figure 4. Hypothetical release curve and hypothetical solubility limits under two conditions A and B with different sump pH or temperatures. The assumed solubility limit for the precipitating phase (precipitate X) is assumed to be 0.6 concentration units under condition A and 0.1 concentration units under condition B.

It is important at this stage to identify the expected precipitates in the post-LOCA sump and to consider the impact of the kinetics on the precipitates formed and their morphology.

Thermodynamic modeling of the ICE tests by McMurry et al. [8] predicted the formation of carbonates, aluminum oxyhydroxide and aluminosilicates, among other species. However, carbonates and aluminosilicates were not observed in the ICE tests, and the model databases had to be modified to improve predictions.

Although silicates were observed in some WCAP tests upon cooling solutions of insulation and concrete dissolution products (both materials contain silicates) exposed to high temperatures (>130°C) and pH extremes, an examination of the kinetics of formation of silicate compounds suggests that in most cases silicate species will not precipitate. Aluminosilicates are complex chemical structures, and their formation from solution is slow at low temperatures. Kinetic effects on the form of precipitates were discussed in detail by McMurry et al. [8], who noted that silicates were unlikely to form in the temperatures and timeframes available in the post-LOCA sump environment. In addition, there is evidence that silicate release can be inhibited by chemical species in the sump water. It was noted by Lane et al. [7] that the release rate of silicates from insulation materials (the primary source) decreases with increasing concentration of aluminum. Such inhibition of silicate release was reported by McMurry et al. [8] whose dissolution tests indicated that silicon release from NuKon[®] was inhibited in the presence of aluminum.

Aluminum hydroxide/oxyhydroxide precipitation was observed in the ICE tests, and the precipitates contained significant amounts of boron. There is significant data in the literature to indicate a strong interaction between boron and aluminum, which can lead to a significant increase (up to a factor of 6) in the solubilities of crystalline aluminum hydroxide (gibbsite and boehmite) [9]. This may be the reason why the corrosion rate of aluminum in borated solutions is much higher than in non-borated solutions of otherwise identical composition; the passive aluminum hydroxide surface film is more soluble in the borated solution. One study of the adsorption of boron onto aluminum hydroxide surfaces found that the boron present in the aluminum hydroxide gel was held predominantly on the surface as a specifically adsorbed ion. Aging studies showed that the adsorption of boron onto the aluminum surface precluded crystallization. Kinetic experiments using pressure-jump-relaxation have also shown that boron adsorbs as an inner-sphere complex on aluminum oxide via a ligand exchange of borate with surface hydroxyl groups [10]. The pzc (point of zero charge) for Al(OH)₃ surfaces is 9.72, whereas for the boron-containing gel, the pzc is in the range of 7.57–8.14. As a result, the presence of boron may significantly change the flocculation behaviour of aluminum hydroxide or oxyhydroxide. Klasky et al. [9] recommended that equilibrium calculations for the ICE test conditions consider the aluminum-borate complex in order to properly assess the effects of the presence of borate ion in the test solution on aluminum solubility. To aid in determining if aluminum hydroxide would precipitate from a borated solution, Bahn et al. [11] presented kinetic stability models based on the results of bench-top tests.

Phase 2 of the AECL approach to chemical effects, bench-top testing, can be performed in cases where precipitation is predicted to be unlikely, in order to provide experimental evidence to support the claim. Bench-top tests may include dissolution tests, where materials are allowed to dissolve under test conditions and the concentrations of relevant chemicals monitored, and/or precipitation tests, where solutions are modified by a change in pH or composition to see if

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precipitation can be induced by a change in chemistry. Tests can also include comparison of the properties of precipitates and surrogates to determine if testing with a surrogate is appropriate.

Phase 3 of the AECL approach to chemical effects, the reduced-scale testing phase, utilizes flow loops with an integrated strainer test module. The industry has developed standard testing methods for the creation of “debris beds”, matted insulation blankets that form under suction when shredded fibrous insulation is introduced; and AECL’s chemical effects testing begins with the creation of a “thin bed”, a mixture of fibrous and particulate insulation displaying a maximum head loss.

In the reduced-scale testing method described by Lane et al. [7] (WCAP-16530), precipitates are formed in concentrated solutions exterior to the test loop, but this method ignores the effects of time, concentration, competing anions and debris bed surfaces on the particle size and distribution of the resulting precipitates. It is unlikely that precipitates formed in this way are representative of precipitates that would form in a post-LOCA environment. The precipitates produced in this way display an amorphous morphology and so-called “prototypical” settling rates; precipitates formed by this method should settle by less than 40% within the first hour of preparation. This is a compromise, meant to reasonably replicate the settling rate observed in Westinghouse tests where solutions from dissolution of aluminum sheet at pH 8 and pH 10 were rapidly cooled. In these tests, precipitates settled by about 30% within 4 hours. Settling rates were found to be a function of the concentration of aluminum oxyhydroxide in the mixing tank before dilution. Solutions with concentrations as high as 11 g/L AlOOH are acceptable in this method. This is an unrealistically high concentration of aluminum, far above the amount of aluminum that could be produced in solution by corrosion of aluminum.

Instead, AECL’s method involves the direct addition of precipitants (soluble chemical precursors such as NaAlO_2 , which hydrolyses to produce $\text{Al}(\text{OH})_4^-$ (aq) within the injected solution, that are expected to form precipitates under the conditions in the test loop). The concentrations of the precipitants only momentarily exceed the sump concentrations predicted from the release equations during injections. This leads to solutions that are far less supersaturated than those produced using the WCAP method, leading to precipitates that are believed to be more representative.

3. THE EFFECTS OF PRECIPITATES ON STRAINER PERFORMANCE

Aluminum and calcium have been identified as the major precipitants in the post-LOCA sump, initially based on the results of the ICE tests and subsequently confirmed by other testing. Precipitates containing primarily aluminum formed in 2 of the 5 Integrated Chemical Effects (ICE) tests [6]. In the WCAP tests, total release of Al from aluminum sheet exposed to pH 4, 8 and 12 at 88 and 130°C exceeded total release of Al, Si, Ca, S, Zn and Fe from fiberglass by a factor of 20-30 and total release from powdered concrete by a factor of 4. Aluminum is present in most nuclear installations as fan blades, scaffolds, CANDU feeder cabinets, and parts of valves and other components. Aluminum precipitates have been observed to produce significant head loss in reduced-scale tests. The Advanced CANDU Reactor (ACR-1000[®]) design has eliminated aluminum from containment.

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Calcium, derived primarily from dissolution of concrete in low-pH solutions ($\text{pH} < 9$) and insulation debris in high-pH solutions ($\text{pH} > 9$), has no known thermodynamically stable precipitates under post-LOCA conditions except in the presence of phosphate [8]. Lane et al. [7] developed a model to calculate calcium release from concrete based on testing where pulverized concrete was exposed to solutions at different pH and temperatures. The model includes an infinitesimal multiplier to convert surface area of exposed concrete to equivalent mass of pulverized concrete, limiting the results of most calculations. In bench-top tests conducted by AECL for Dominion Generation, low-grade concrete was found to dissolve readily below pH 8, but in the presence of TSP dissolution was almost completely inhibited. This may have been the result of the formation of a protective calcium phosphate surface film on the concrete. Furthermore, in reduced-scale chemical-effects tests for Dominion Generation, additions of calcium chloride had a negligible effect on strainer head loss. Therefore, the importance of calcium in the analysis of chemical effects on strainer performance may be called into question. However, it may be noted as a point of interest, Scanning Electron Microscopy (SEM) analysis of debris bed fibers in tests where calcium was added showed indications of a Ca-Al-P precipitate, and calcium concentration was observed to decrease in a one-to-one molar ratio with aluminum additions (Figure 5). Thermodynamic analysis indicated that $\text{CaAlH}(\text{PO}_4)_2$ may form under certain conditions, but has been reported to be unstable with respect to hydrolysis [12]. Calcium was noted to precipitate in the absence of significant dissolved aluminum once the concentration exceeded about 24 mg/L Ca; this concentration exceeds the solubility of $\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$ by a factor of 4 and $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ by a factor of 30 billion, but it is not clear which compound may have precipitated. This precipitation was not observed to increase head loss.

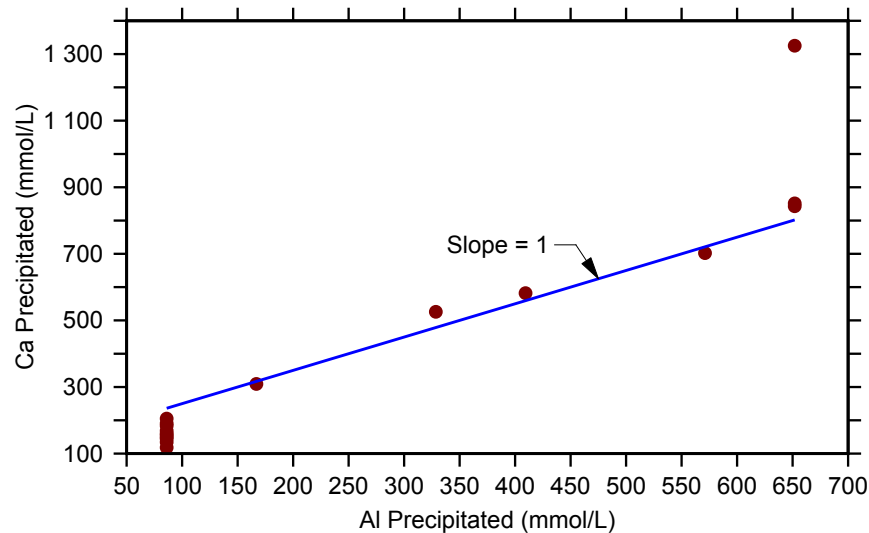


Figure 5. Quantity of precipitated calcium as a function of precipitated aluminum. Calcium and aluminum appear to have co-precipitated in the presence of phosphate. (Dominion Generation reduced-scale chemical effects test data. Reproduced with permission.)

Additions of sodium aluminate (NaAlO_2) solutions have been observed to result in increases in pressure drop (head loss) across strainers covered with thin beds of fibrous insulation. Thin beds represent the most conservative condition for testing and are created by adding fiber to loops

containing particulate debris; the fiber mixes with the particulate, resulting in a very dense and rather impermeable mat. Additions were not observed to result in homogeneous precipitation—samples taken downstream of the addition point did not contain precipitates or significant turbidity—but aluminum concentrations seldom exceeded the detection limit for ICP-AES (Inductively Coupled Plasma Atomic Emission Spectroscopy) (0.4 mg/L), suggesting rather quick and complete precipitation of aluminum. SEM examination of debris beds showed primarily uniform distribution of precipitates, suggesting that heterogeneous precipitation on insulation fibers was the primary precipitation mechanism. This was not unexpected since heterogeneous precipitation was also observed in the ICE tests [13]. As noted by Bahn et al. [13], heterogeneous nucleation is favored over homogeneous nucleation if interactions between a nucleating particle and a surface lower the net interfacial energy more than the interactions between the particle and the solution. It is expected that precipitation within the debris bed results in tightening or clogging of pores in the debris bed, resulting in head loss increases.

Figure 6 shows a representative head loss curve from a test conducted by AECL for Dominion Generation in borated water at pH 7 and 40°C. Frequently, head loss peaked after additions, only to stabilize to a lower value. The peak head losses are plotted against the amount of aluminum precipitated per unit area of strainer (the strainer aluminum load) in Figure 7. Using the available pump suction head margin, the maximum allowable strainer aluminum load can be calculated and used to justify the existing aluminum components in containment or their replacement.

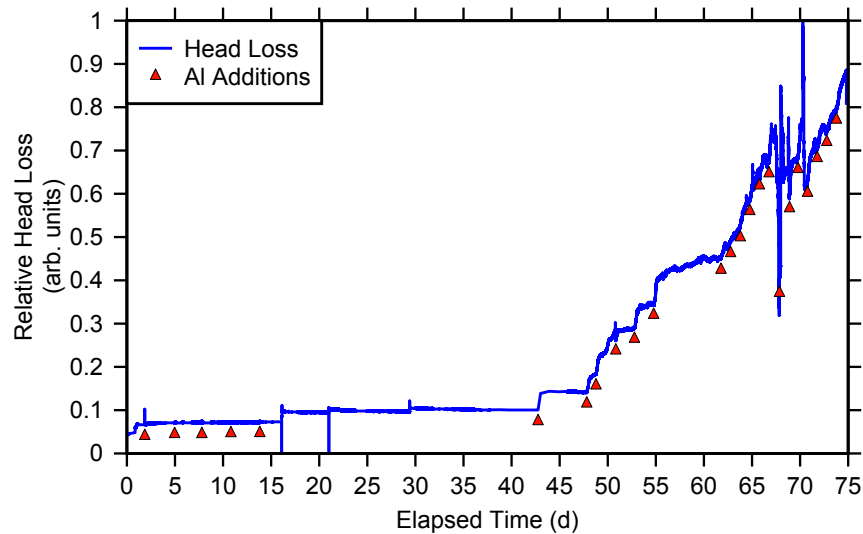


Figure 6. Head loss observed during a typical chemical effects test. Aluminum additions in the form of dissolved NaAlO_2 were observed to increase head loss significantly, often resulting in head loss peaks. (Dominion Generation reduced-scale chemical effects test data. Reproduced with permission.)

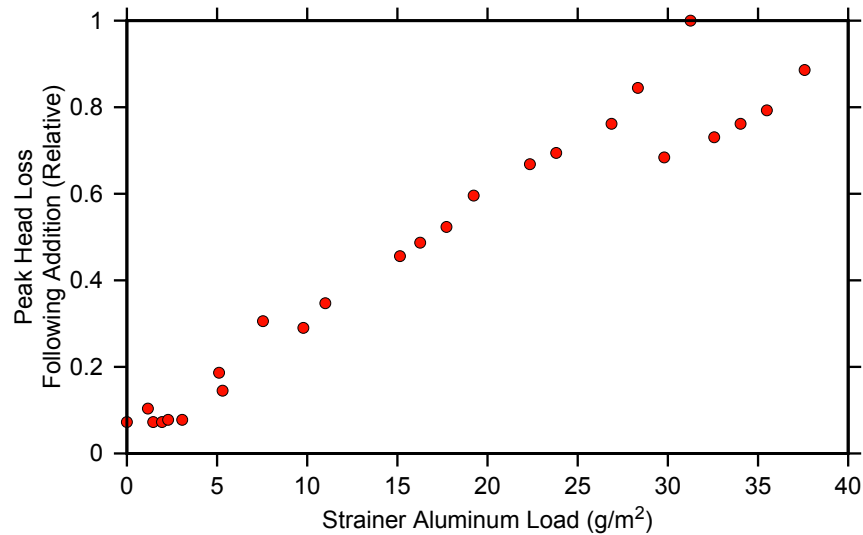


Figure 7. Peak head loss was observed to increase almost linearly with the amount of precipitated aluminum per unit area of strainer (strainer aluminum load). (Dominion Generation reduced-scale chemical effects tests data. Reproduced with permission.)

4. PREDICTING CHEMICAL EFFECTS HEAD LOSS

The AECL *Finned Strainer* incorporates very large surface areas into a compact design. As such, flow is typically laminar, even under thin-bed conditions. If sufficient quantities of precipitates form on thin beds, however, turbulent conditions are expected to be created. The debris bed may be modeled as an array of pores through which most of the fluid passes. As precipitates form within the debris bed, the pores get clogged, increasing pressure drop across the strainer. In such a model, the fluid velocity through remaining pores increases as precipitants are added and precipitates form until turbulent conditions exist within those pores, at which point further increases in head loss are not expected since fluid shear would prevent precipitates from adsorbing or else cause them to spall. This may help to explain the head loss plateau observed in many tests, where the addition of more aluminum did not result in significant head loss increases.

There is a clear relationship observed when chemical effects test data are modeled after an orifice. The flow equation for an orifice is given by Equation (4).

$$Q = C_{discharge} A_{orifice} \sqrt{2g\Delta h} \quad (4)$$

where Q is the volumetric flow rate, $C_{discharge}$ is the discharge coefficient, $A_{orifice}$ is the cross-sectional area of the orifice or equivalent orifice area resulting from many debris bed pores, g is the gravitational constant and Δh is the head loss across the orifice or strainer. The calculated $A_{orifice}$ with an assumed nominal value of $C_{discharge} = 1$, normalized to the size of the strainer, is shown in Figure 8. A clear limit is reached in many tests where further increases in aluminum load do not increase the head loss or equivalent orifice area.

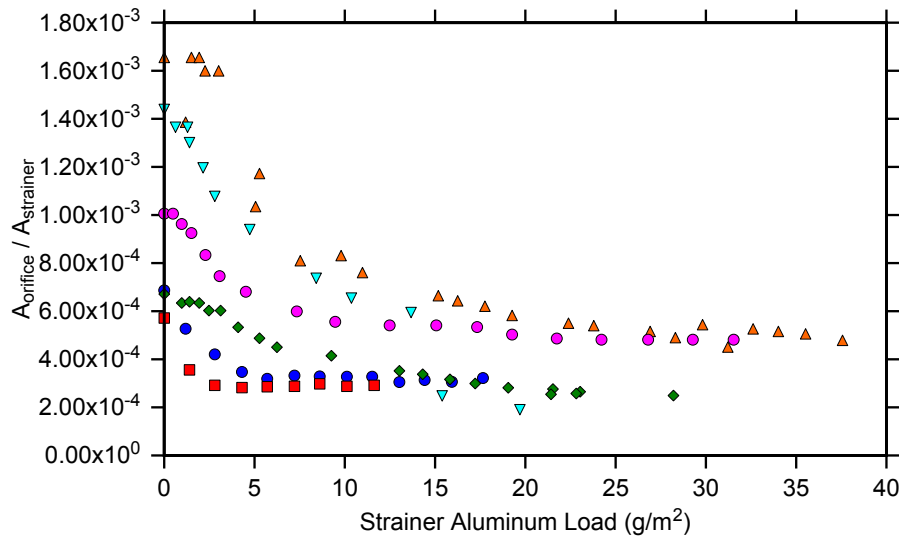


Figure 8. Chemical effects head loss data modeled after an orifice (Equation 4). Pores in the debris bed are equated to an orifice area and plotted against the strainer aluminum load. (Dominion Generation reduced-scale chemical effects tests data. Reproduced with permission.)

At low aluminum loads the debris bed is more porous and laminar conditions exist. A transition to turbulent flow occurs at some critical aluminum load; at the end of the tests shown in Figure 8 the head loss was found to be proportional to the flow rate to the power of about 1.8 (an exponent of 1 is expected for laminar flow and 2 for turbulent flow).

While few of the data sets agree, a reasonable estimate of worst-case head loss could be calculated using $A_{\text{orifice}}/A_{\text{strainer}} = 2 \times 10^{-4}$. Of course, there is no substitute for testing, as each combination of strainer size, debris types and quantities, as well as quantities of precipitants, produces a unique result. New *Finned Strainer* designs, which incorporate further increases in surface area density, should experience lower aluminum loads and thus lower head loss.

5. SUMMARY

The three-phase AECL approach to evaluating the effects of sump water chemistry on ECCS strainer performance involves:

- 1) careful consideration of available literature and release (corrosion and dissolution) models and evaluation of sump environments;
- 2) bench-top testing to provide data for release and precipitation models, to gain a better understanding of sump chemistry, and/or to determine if testing with a surrogate is appropriate; and,
- 3) reduced-scale chemical-effects testing to provide head loss data.

AECL has developed a unique chemical-effects testing method whereby precipitants (such as $\text{Al}(\text{OH})_4^-(\text{aq})$) are added to a closed test loop with simulated sump water (e.g., added boric acid) and long-term sump temperatures. In this method, precipitates form within the debris bed in a representative sump environment. This method differs from the method proposed in

WCAP-16530, in which precipitates are formed in highly concentrated solutions exterior to the test loop and are added like another form of debris.

Chemical-effects testing has shown a clear relationship between the amount of aluminum precipitated on strainer debris beds and strainer head loss. Conversely, calcium was not observed to precipitate readily, and was only observed to increase head loss when co-precipitating with aluminum. The importance of calcium in the analysis of chemical effects on strainer performance is usually not a significant factor. Reduced-scale chemical-effects test data may be used to determine the maximum allowable mass of precipitated aluminum on a strainer design and, using post-LOCA sump conditions and an aluminum release model, calculate the mass of aluminum release to justify the existing aluminum components in containment or their replacement.

6. ACKNOWLEDGEMENTS

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