



Printed in the United States of America. Available from  
the Energy Research and Development Administration,  
Technical Information Center  
P.O. Box 62, Oak Ridge, Tennessee 37830  
Price: Printed Copy \$5.50 ; Microfiche \$2.25

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ORNL-TM-5064  
UC-79c - Fuel Recycle

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

FIXATION OF RADIOIODINE WITH PORTLAND CEMENT. I. PRELIMINARY SCOPING  
STUDIES

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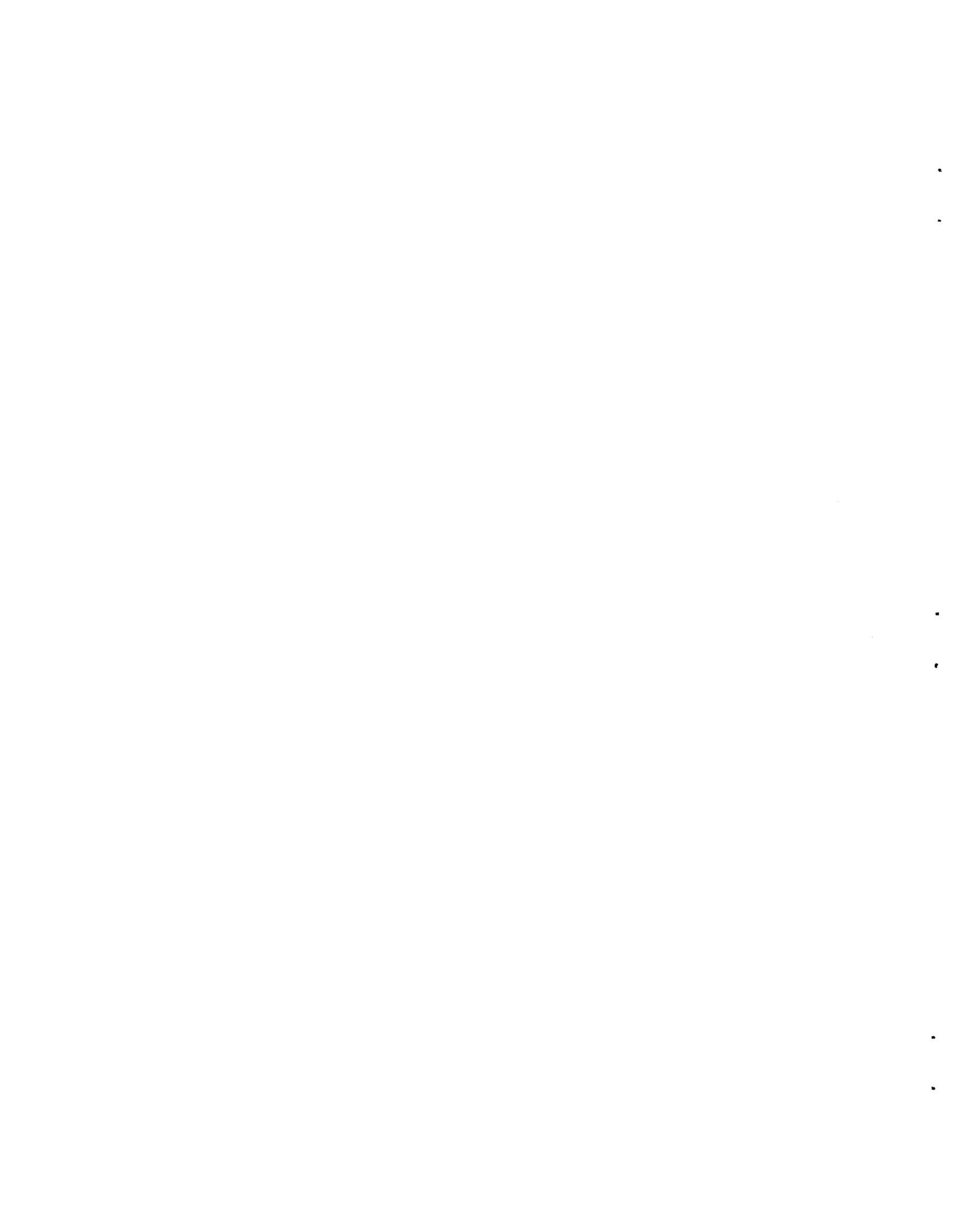
DECEMBER 1975



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## FIXATION OF RADIOIODINE WITH PORTLAND CEMENT. I. PRELIMINARY SCOPING STUDIES

W. E. Clark  
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## ABSTRACT

Data are presented for the short-term ( $\leq 7$  months) leaching of iodine from concrete specimens containing 2.9, 5.7, 7.5, and 9.05 wt % iodine as barium iodate. In this study, which is still in progress, the incremental leach rates of specimens leached for 205 days have decreased to less than  $10^{-4}$  cm/day. Those containing 9.05 wt % iodine have been leached for much shorter periods, and only the best specimens (i.e., those containing either butyl stearate or fluosilicic acid as additives) show leach rates as low as  $10^{-4}$  cm/day.

Fixation of radioiodine as barium iodate in concrete offers possibilities for actual disposal. The product, if prepared free of all radionuclides except iodine, is a low-level waste and can be transported and stored like other solid low-level wastes. The total daily iodine output of a 5-metric ton/day LMFBR fuel reprocessing plant can be contained in only  $0.335 \text{ ft}^3$  of concrete. Heat generation in a product containing 9.05 wt % iodine would amount to only  $3.3 \times 10^{-3}$  W per metric ton of concrete.

Recommendations are made for further work toward optimizing the product.

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## 1. INTRODUCTION

The traditional method for handling radioiodine from reactor and fuel processing off-gas has been to temporarily remove it from the off-gas, usually by adsorption on a solid sorbent. The iodine thus removed is held up long enough for the short-lived activities to decay and is then desorbed and released to the environment. Since all of the radioisotopes except I-129 are relatively short-lived (Table 1) and since I-129 has a relatively low fission yield as well as a low specific activity, this has been considered to be acceptable practice until recently. However, the long half-life of I-129 ( $1.59 \times 10^7$  years) ensures that essentially all of the isotope produced will persist throughout foreseeable human existence. While

Table 1. The more stable isotopes of iodine and their characteristics<sup>a,b</sup>

| Mass<br>number | Half-life                | Radiation                                    |  | Disintegration<br>energy (MeV)                 |
|----------------|--------------------------|--|--|--|
|                |                          | Type <sup>c</sup>                            | Energy (MeV)                               |  |
| 124            | 4.17 d                   | $\epsilon, \beta^+$<br>$\gamma$              | 1.54, 2.14<br>6.03 to 2.988                | 3.16   |
| 125            | 59.7 d                   | $\epsilon$<br>$\gamma$                       | 0.035                                      | 1.48   |
| 126            | 13.0 d                   | $\epsilon, \beta^+$<br>$\beta^-$<br>$\gamma$ | 1.13, 0.46<br>0.86, 1.25<br>0.389 to 2.045 | 2.15(E <sup>+</sup> )<br>1.25(E <sup>-</sup> ) |
| 127            | Stable                   |  | -  | -  |
| 129            | 1.59 x 10 <sup>7</sup> y | $\beta^-$<br>$\gamma$                        | 0.15<br>0.0396                             | 0.19   |
| 131            | 8.041 d                  | $\beta^-$<br>$\gamma$                        | 0.606 to 0.81<br>0.36 to 0.802             | 0.971  |

<sup>a</sup>Includes all known isotopes of iodine with  $t_{1/2} \geq 24$  hr.

<sup>b</sup>Data condensed from "Chart of the Nuclides," Knolls Atomic Power Laboratory, April 1972.

<sup>c</sup> $\epsilon$  = electron capture.

the radiation level of I-129 justifies its classification as a low-level waste, the potential for accumulation in the biosphere requires that it be permanently sequestered so as to prevent such an accumulation. The simplest and most obvious method seems to be conversion to some stable, insoluble, solid compound of iodine which can act as a filler in the formation of concrete from Portland cement or some other stable and insoluble, solid material. The resulting product should be suitable for storage in any facility approved for the containment of low-level radioactive waste.

Cement has been used for more than two decades to immobilize solid fission products which have half-lives too long to allow their decay and release but are present in quantities small enough that heat generation does not adversely affect the integrity of the resulting concrete.\* Generally, the fission product in question is first converted into a relatively insoluble form either by precipitation as an insoluble compound or, as in the case of Cs-137, by adsorption on some inert solid (e.g., grundyte clay for cesium) which is subsequently incorporated into the concrete.<sup>1</sup> It is expected that iodine and other radioactive wastes will be stored in such a manner that they are kept dry in perpetuity. The insolubility of the product becomes an added safety factor in case of unforeseen accidents.

The objective of the work reported here is to develop concrete compositions which contain substantial percentages of iodine while retaining good resistance to leaching and satisfactory mechanical strength. The purpose of this paper is to report the results of the first preliminary experiments in the development of such compositions. Our immediate goal in these preliminary studies is to determine the applicability of the general method for immobilizing iodine from reprocessing schemes which generate iodate waste (e.g., the Iodex process<sup>2</sup>) and to establish a base of experimental data for future work.

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\*In this report, the term "cement" refers to the commercial dry material and "cement paste" to the solid product resulting from the addition of water only. The term "concrete" denotes the final product resulting from the addition of iodine compounds and other materials to the cement-water mixture.

The work is still in its early stages, and leaching tests on the concrete specimens now under way should be continued for several more months. However, the possibility that we will be unable to continue the program to its logical conclusion makes it desirable that we report the results obtained thus far, even though they are incomplete.

## 2. GENERAL CONSIDERATIONS

Iodine forms relatively few stable and insoluble, simple inorganic compounds such as would be preferred for long-term storage in cement (Table 2). Among the iodides, the silver and monovalent mercury salts are, by far, the most insoluble; however, the cost of silver and mercury makes such compounds economically unattractive if satisfactory alternatives can be found. Lead iodide, while much more soluble, may be an acceptable substitute. Among iodates, the mercurous salt is again the most insoluble. Lead, silver, and barium compounds are the next most attractive, although all are more soluble than would be ideal for this application. The use of either barium iodate or lead iodate is somewhat complicated by the fact that the sulfates of both metals are more insoluble than the iodates. Type 1 cement usually contains 2 to 3%  $\text{SO}_3$ , which may cause conversion of the iodates of barium or lead to the more soluble calcium salt (Table 2).

Barium iodate is known to be thermally stable to at least  $476^\circ\text{C}$ , except for loss of water from the monohydrate.<sup>3</sup> Even  $\text{HI}_3\text{O}_8$ , the partially dehydrated acid, is thermally stable to about  $229^\circ\text{C}$ .<sup>4</sup> Lead iodate is reported to decompose at about  $300^\circ\text{C}$ .<sup>5</sup> In any case, thermal stability at temperatures greatly in excess of  $100^\circ\text{C}$  is not a major criterion for disposal of iodine in concrete.

Our first criterion is that a waste disposal scheme should make use of the chemical form most compatible with the process that generates the waste unless there are overriding considerations to the contrary. In our case, the process under development (Iodox) produces a partially dehydrated form of iodic acid,  $\text{HI}_3\text{O}_8$ , by precipitation from 20 M  $\text{HNO}_3$ . It is, therefore, more expedient for us to consider incorporation as

Table 2. Solubilities of selected iodine compounds<sup>a</sup>

| Cation    | Solubilities <sup>b</sup> (g-moles/liter at 25°C) <sup>c</sup> |  |
|-----------|--|--|
|           | Iodide   | Iodate   |
| Sodium    | 8.2  | 0.47   |
| Potassium | 4.7  | 0.42   |
| Magnesium | 5 molal <sup>d</sup>   | 0.25   |
| Calcium   | 4.9 at 20°C  | $7.9 \times 10^{-3}$   |
| Barium    | 4.0  | $8.1 \times 10^{-4}$   |
| Lead      | $1.65 \times 10^{-3}$  | $(3.6 \text{ to } 5.5) \times 10^{-5}$                         |
| Silver    | $1.1 \times 10^{-8}$   | $1.8 \times 10^{-4}$   |
| Mercuric  | $9.7 \times 10^{-5}$ to $1.3 \times 10^{-4}$                   | "Insoluble" <sup>e</sup>                                       |
| Mercurous | $3 \times 10^{-10}$ (Hg <sub>2</sub> I <sub>2</sub> )          | $(1.1 \text{ to } 1.6) \times 10^{-9}$<br>(HgIO <sub>3</sub> ) |

<sup>a</sup>The cations selected include those most abundant in Portland cement (Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>; omitting Al<sup>3+</sup>, Fe<sup>3+</sup>, and Si<sup>4+</sup>) and those which form the most insoluble, simple compounds of iodine (Ba<sup>2+</sup>, Ag<sup>+</sup>, Pb<sup>2+</sup>, Hg<sup>2+</sup>, and Hg<sub>2</sub><sup>2+</sup>).

<sup>b</sup>Molar solubilities were calculated from data obtained from A. Seidell and W. F. Linke, Solubilities of Inorganic and Metal-Organic Compounds, 4th ed., American Chemical Society, 1958, except as indicated.

<sup>c</sup>In all cases except where indicated.

<sup>d</sup>Lack of solution density data prevented conversion to molarity.

<sup>e</sup>N. A. Lange, Handbook of Chemistry, Rev. 10th ed., McGraw-Hill, New York, 1967.

iodate than as iodide. Iodide, a reductant, is easily oxidized to elemental iodine, sometimes even by air; iodate, an oxidant, can be readily reduced to the elemental form. Both iodide and iodate will produce free iodine when irradiated in air. Since concrete is neither oxidizing nor reducing, either iodate or iodide is compatible with this solidification medium; however, elemental iodine will doubtless be generated by radiolysis during long-term storage in either form. The long half-life and weak radiation from I-129 allow us to consider storage in concrete in spite of the possibility that iodine will be generated very slowly.

A second criterion for our process is that the preliminary treatment which results in the formation of the insoluble iodate should involve the addition of as few soluble ions as possible. This suggests the use of oxides or hydroxides in preparing the insoluble iodate. Since the final process should be as simple as possible, we plan to generate the insoluble iodate by using volumes of water which can be completely incorporated in the concrete product. In this way, no aqueous waste stream will be generated from the precipitation process.

A third criterion of any practical waste disposal project is to reduce the volume to be disposed of to as low a value as possible. We should, therefore, attempt to incorporate as much iodine as possible into the final product, consistent with other constraints such as mechanical strength of concrete, internal heat generation, leach rates, etc.

The iodic acid resulting from the Iodex process is essentially 100%  $\text{HI}_3\text{O}_8$ . It is estimated<sup>6</sup> that the fission product iodine after the holding tank will consist of 75.5% I-129, with the remainder being stable I-127. If this is incorporated in concrete to the extent of 10 wt % iodine, the heat generation will amount to about  $3.6 \times 10^{-9}$  W/g or  $3.6 \times 10^{-3}$  W/metric ton.

Even though the most important items in the quality of a standard commercial concrete are the care with which the aggregate is mixed and the effectiveness of the curing process used, it may be desirable to add special waterproofing and/or dampproofing agents to obtain the

lowest possible leach rates. Numerous materials have been tried for their effectiveness in this regard. Those generally considered<sup>7-9</sup> as the most effective include entrained air, gas-forming agents such as metallic aluminum powder, calcium chloride, soaps, butyl stearate, finely divided dry materials (e.g., pozzolans), heavy mineral oil, and various agents which increase the workability of the cement. Some materials reputedly used in proprietary additives such as fluosilicic acid and various compounds of iron, aluminum, and copper were not specifically mentioned in any references we found.

As a matter of principle, we would prefer to keep the concrete free of organic materials since there is always some possibility that labile organic iodides (e.g., methyl iodide) may be formed via the reduction of iodate in the presence of organic material and radiation. This effect may, however, be unimportant in our process because of the very low energy of the radiation from the decay of I-129 and the high degree of internal shielding supplied by the concrete matrix.

### 3. EXPERIMENTAL

We converted reagent-grade iodic acid,  $\text{HIO}_3$ ,\* to barium iodate by reacting the dissolved acid with a slight stoichiometric excess of aqueous barium hydroxide so that the resulting slurry was always alkaline. Volumes of water were chosen such that the resulting slurry supplied all the water needed for making up the concrete aggregate. The excess hydroxyl ion, although undesirable, was considered to be more acceptable than any available alternative.

We also attempted to convert lead oxide ( $\text{PbO}$ ) to the iodate by direct reaction with iodic acid, but the reaction was unacceptably slow at room temperature. Incorporation of the unreacted  $\text{PbO-HIO}_3$  slurry into concrete resulted in overheating as the unreacted acid attacked the calcined metal oxides in the cement. Therefore, we abandoned the idea of using lead iodate and concentrated on barium iodate instead.

---

\*We believe that the behavior of  $\text{HIO}_3$  will be equivalent to that of  $\text{HI}_3\text{O}_8 + \text{H}_2\text{O}$ . This must, however, be confirmed.

Specimens containing between 2.9 and 9.05 wt % iodine were made up by mixing in a Waring-type blender at speeds which started at about 1000 rpm and were increased to about 3000 rpm toward the end of the mixing process. Type 1 cement<sup>10</sup> was used in all cases. The cement was then poured into 5-cm-diam by 5-cm-high cylindrical polyethylene containers in order to conform with the specifications of the International Atomic Energy Agency (IAEA).<sup>11</sup> After casting, the samples were allowed to stand in a water-saturated atmosphere for 28 days or longer.

When great care was taken to remove the air bubbles from the cement, the samples filled the container snugly, leaving no visible void between the container wall and the cement. Unless such care was taken and the specimens were properly cured in water-saturated air, the leaching results were sufficiently erratic to overshadow the effect of varying the iodine content of the concrete. It was, in fact, necessary to discard the first batch of specimens cast because of excessively poor reproducibility of the initial leach results.

Initially, three series (Series A, B, and C; see Table 3) of specimens were prepared and leached in triplicate with distilled water (boiled to remove dissolved CO<sub>2</sub>). Our objectives were: (1) to determine the effect of varying the content of barium iodate (2.9 to 7.5 wt % iodine) while maintaining an essentially constant cement-to-water ratio, and (2) to determine the degree of reproducibility that can be expected between specimens. Each leach specimen was made up independently from the starting materials and mixed individually; that is, specimens in a series were not simply different aliquots of the same mix.

The next six series of specimens (Series D, E, F, G, H, and I; see Table 3) were designed to test the effect of increasing the iodine content still further (to 9.05 wt % iodine) and to test the effect of various additives on the leaching rate of this particular composition. Leaching tests on these specimens were run in duplicate.

Table 3. Compositions of leached specimens

| Specimen designation | Component (wt %) <sup>a</sup> |                    |     |                               |                                       | Total | Percent iodine |
|----------------------|-------------------------------|--------------------|-----|-------------------------------|---------------------------------------|-------|----------------|
|                      | Cement                        | Water <sup>b</sup> | BaO | I <sub>2</sub> O <sub>5</sub> | Additives                             |       |                |
| A-1<br>-2<br>-3      | 46.7                          | 38.9               | 4.5 | 9.9                           | None                                  | 100.0 | 7.5            |
| B-1<br>-2<br>-3      | 53.7                          | 40.8               | 1.7 | 3.8                           | None                                  | 100.0 | 2.9            |
| C-1<br>-2<br>-3      | 50.0                          | 39.8               | 3.2 | 7.0                           | None                                  | 100.0 | 5.4            |
| D-1<br>-2            | 44.2                          | 38.3               | 5.6 | 11.9                          | None                                  | 100.0 | 9.05           |
| E-1<br>-2            | 44.2                          | 37.8               | 5.6 | 11.9                          | Na <sub>2</sub> SiO <sub>3</sub> -0.5 | 100.0 | 9.05           |
| F-1<br>-2            | 44.2                          | 37.8               | 5.6 | 11.9                          | H <sub>2</sub> SiF <sub>6</sub> -0.5  | 100.0 | 9.05           |
| G-1<br>-2            | 44.2                          | 37.3               | 5.6 | 11.9                          | Butyl stearate-0.9                    | 99.9  | 9.05           |
| H-1<br>-2            | 44.1                          | 36.8               | 5.6 | 11.9                          | Polybutene 24-1.2;<br>Aerosol OT-0.5  | 100.1 | 9.05           |
| I-1<br>-2            | 44.2                          | 36.9               | 5.6 | 11.9                          | PbO-1.4                               | 100.0 | 9.05           |

<sup>a</sup>For simplicity in reporting, barium iodate is broken down into the elemental oxides [Ba(IO<sub>3</sub>)<sub>2</sub> = BaO + I<sub>2</sub>O<sub>5</sub>]. Ba(OH)<sub>2</sub>·8H<sub>2</sub>O was reacted with HIO<sub>3</sub> to form the barium iodate actually used.

<sup>b</sup>Total water, including that from the reaction of Ba(OH)<sub>2</sub>·8H<sub>2</sub>O and HIO<sub>3</sub> as well as that added.

Static leaching tests were run under a minimum water depth of 5 cm. In the containers used, the volume of distilled water required for leaching was 300 ml. These tests were conducted in accordance with the IAEA recommended procedures,<sup>11</sup> except that:

1. Sampling and leachant change were done on a daily basis for the first three weeks, except on weekends and holidays. Manpower and budgetary requirements dictated this variation.
2. Daily sampling and leachant change were discontinued on Series A, B, and C when, after about three weeks, it appeared (erroneously) that the method of analysis used would not be sufficiently sensitive to determine the iodine in the leachant over such short leaching periods. In the later series (D through I), daily sampling and leachant change were continued for about five weeks. The IAEA requirement that the pH must be below 8 before daily sampling is discontinued was particularly impractical in our case since excess barium hydroxide was present in all specimens and the pH has remained high throughout the tests. In all cases, the pH of the leachant was still greater than 10 at the time that daily sampling was discontinued.

Our deviation from the regimen of daily sampling caused marked discontinuities in some of the data plots (Figs. 1-12) until we adopted the regimen of sampling once weekly.\*

We used a conventional spectrophotometric method of analysis instead of employing a radioisotope of iodine. The isotopes, except for I-129 and possibly I-125, have half-lives much too short to be suitable as tracers in long-term tests (Table 1). Unfortunately, the low specific activity of I-129 and the softness of the radiation from both I-129 and I-125 make the utility of either of these isotopes marginal at best. Activation analysis is too expensive and time-consuming for our application. On the other hand, the use of iodate as the fixation form for iodine allows us to multiply the effective concentration of iodine in the leachate by a factor of 6.

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\* Some of these discontinuities were too large to be explained by a change in sampling regimen, as will be explained later.

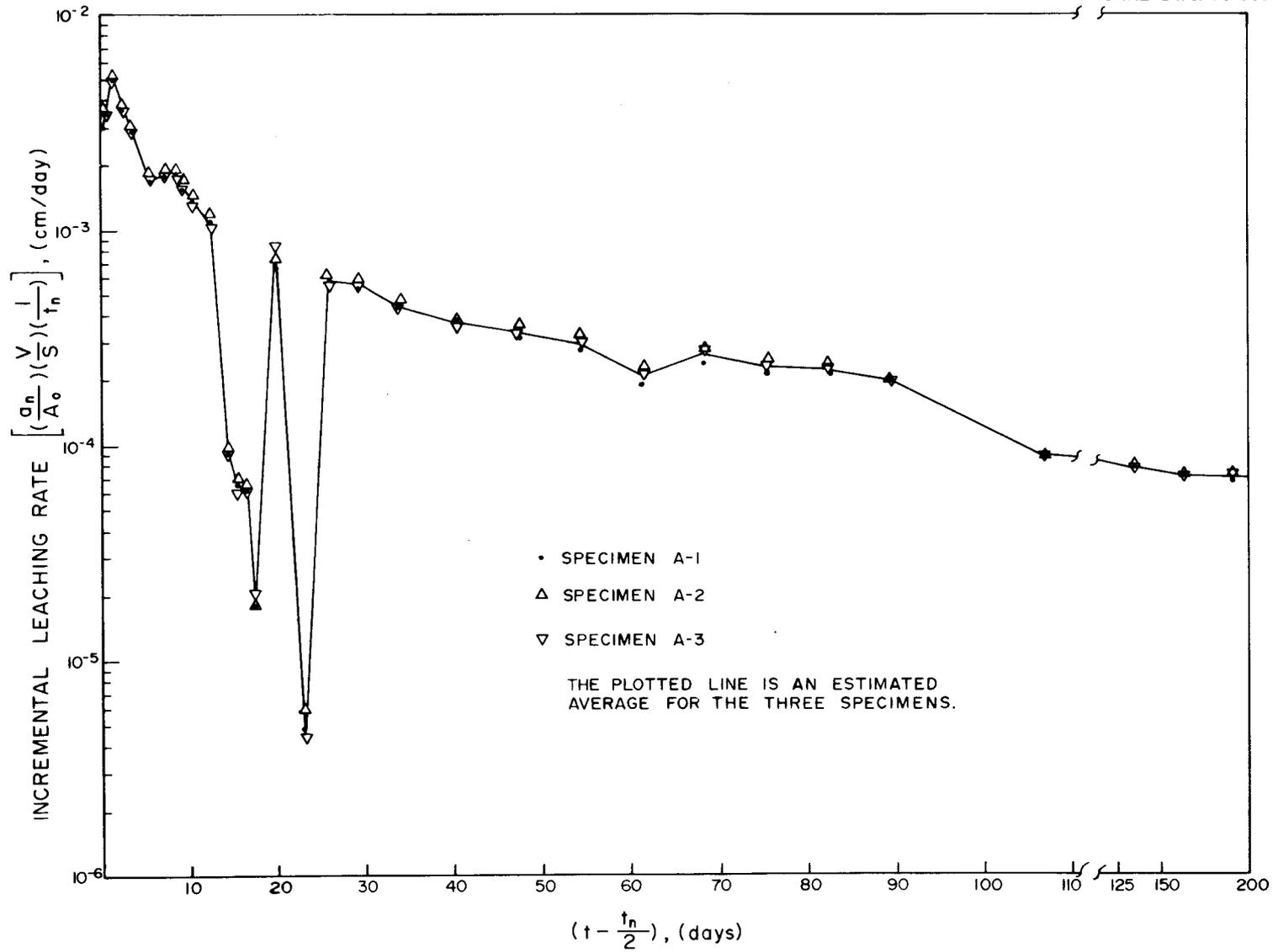


Fig. 1. Incremental leaching rates for Specimens A-1, A-2, and A-3, which contain 7.5% iodine as barium iodate. See Tables A-1, A-2, and A-3 for raw data.

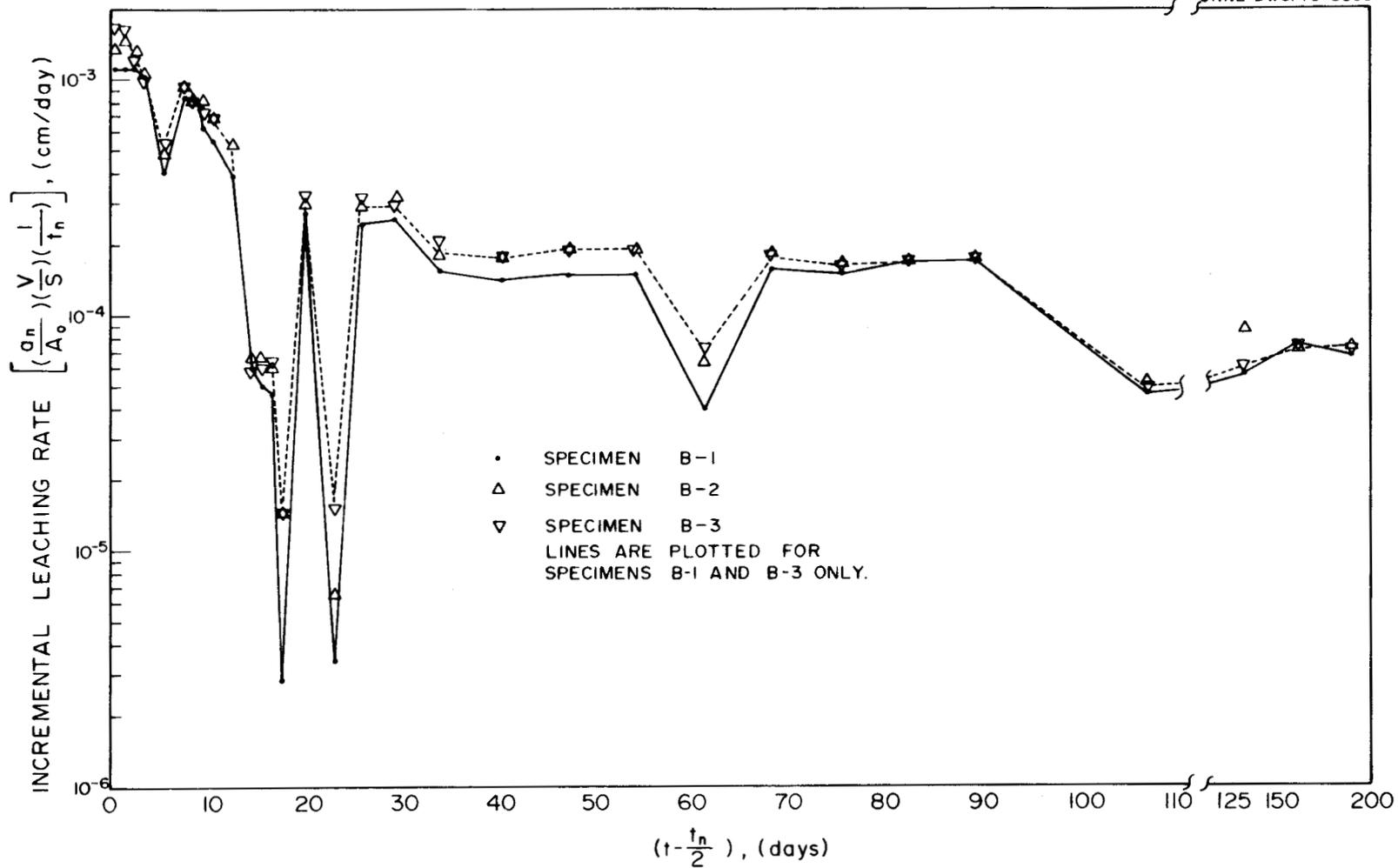


Fig. 2. Incremental leaching rates for Specimens B-1, B-2, and B-3, which contain 2.9% iodine as barium iodate. See Tables A-4, A-5, and A-6 for raw data.

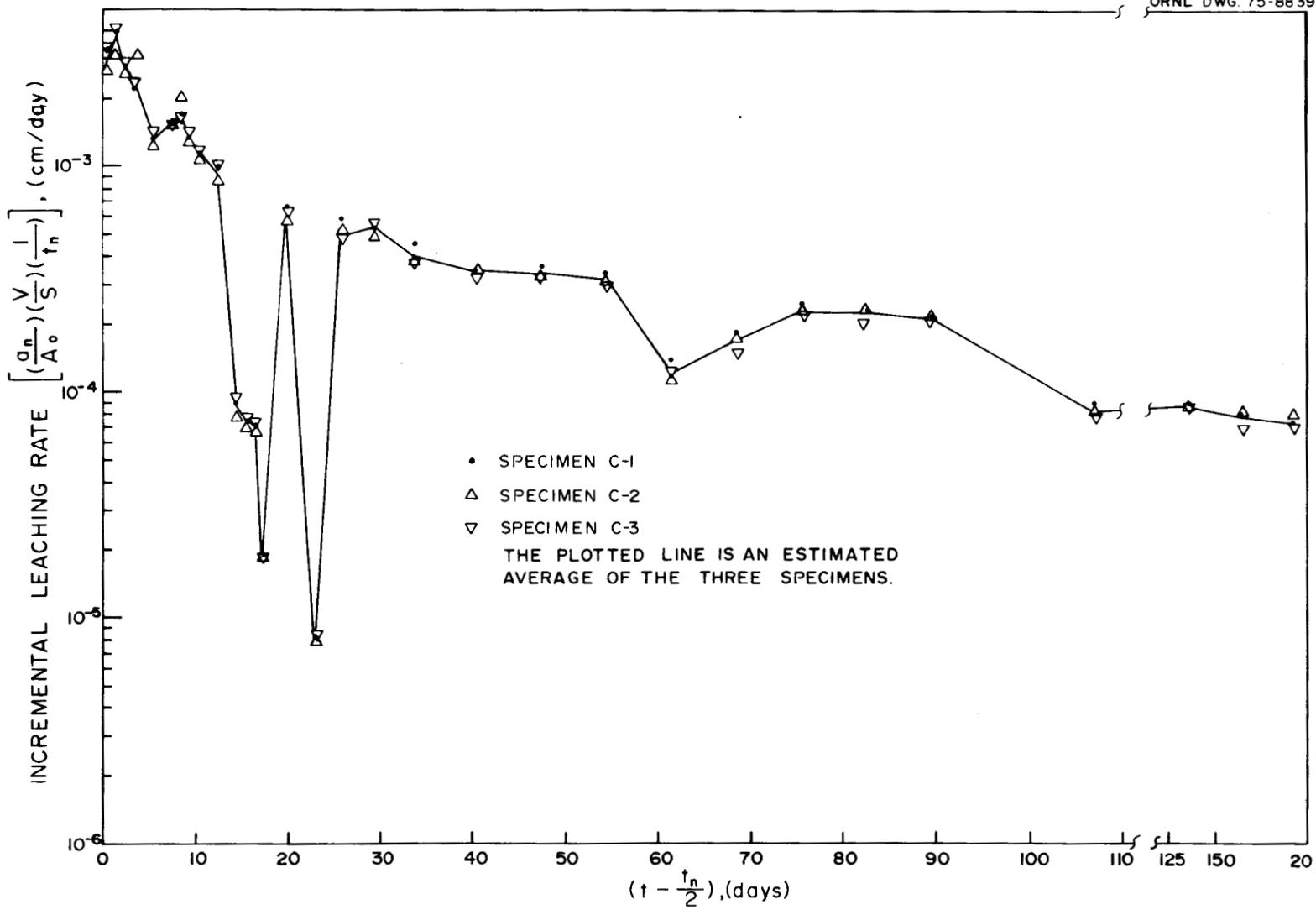


Fig. 3. Incremental leaching rates for Specimens C-1, C-2, and C-3, which contain 5.4% iodine as barium iodate. See Tables A-7, A-8, and A-9 for raw data.

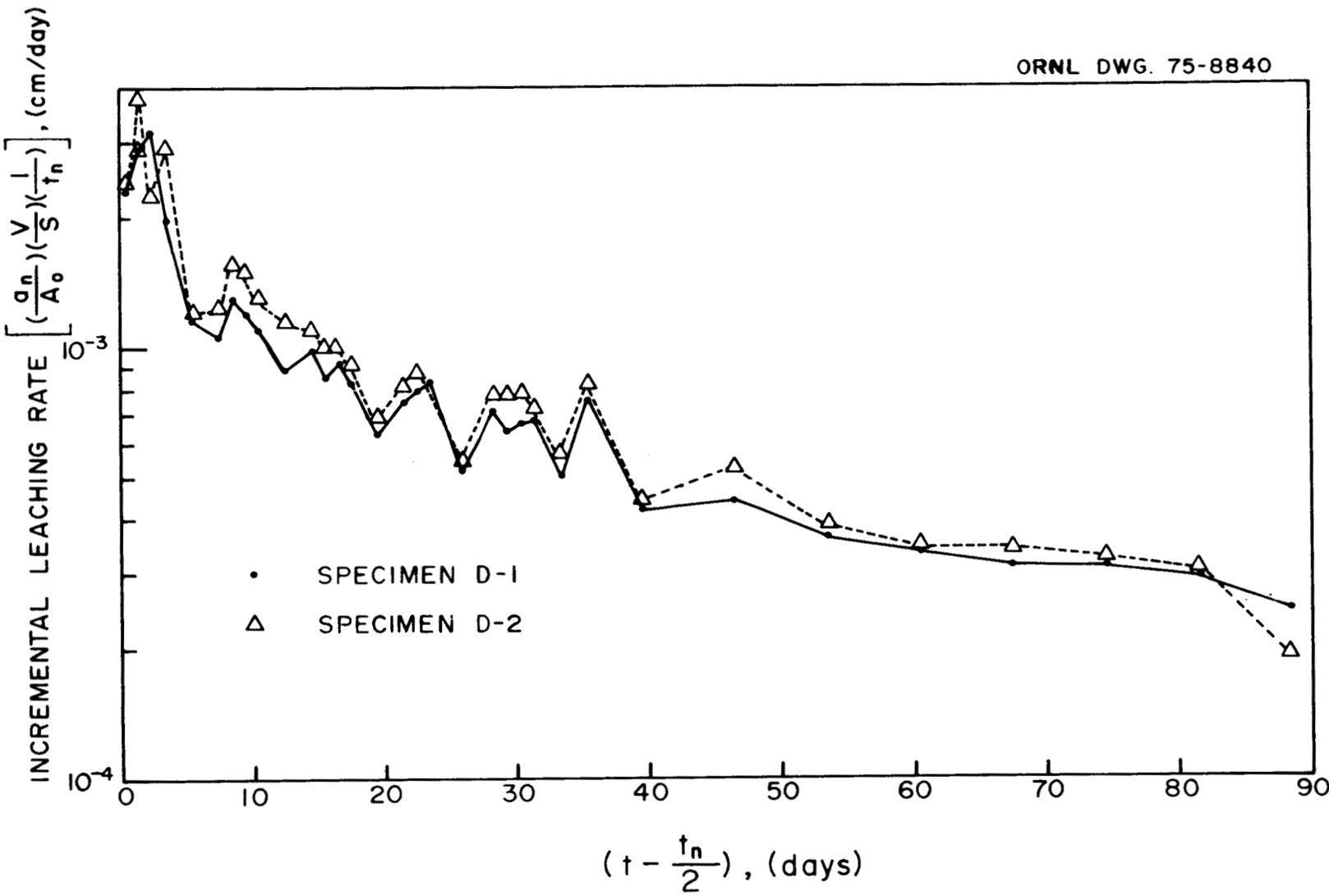


Fig. 4. Incremental leaching rates for Specimens D-1 and D-2, which contain 9.05% iodine as barium iodate. See Tables A-10 and A-11 for raw data.

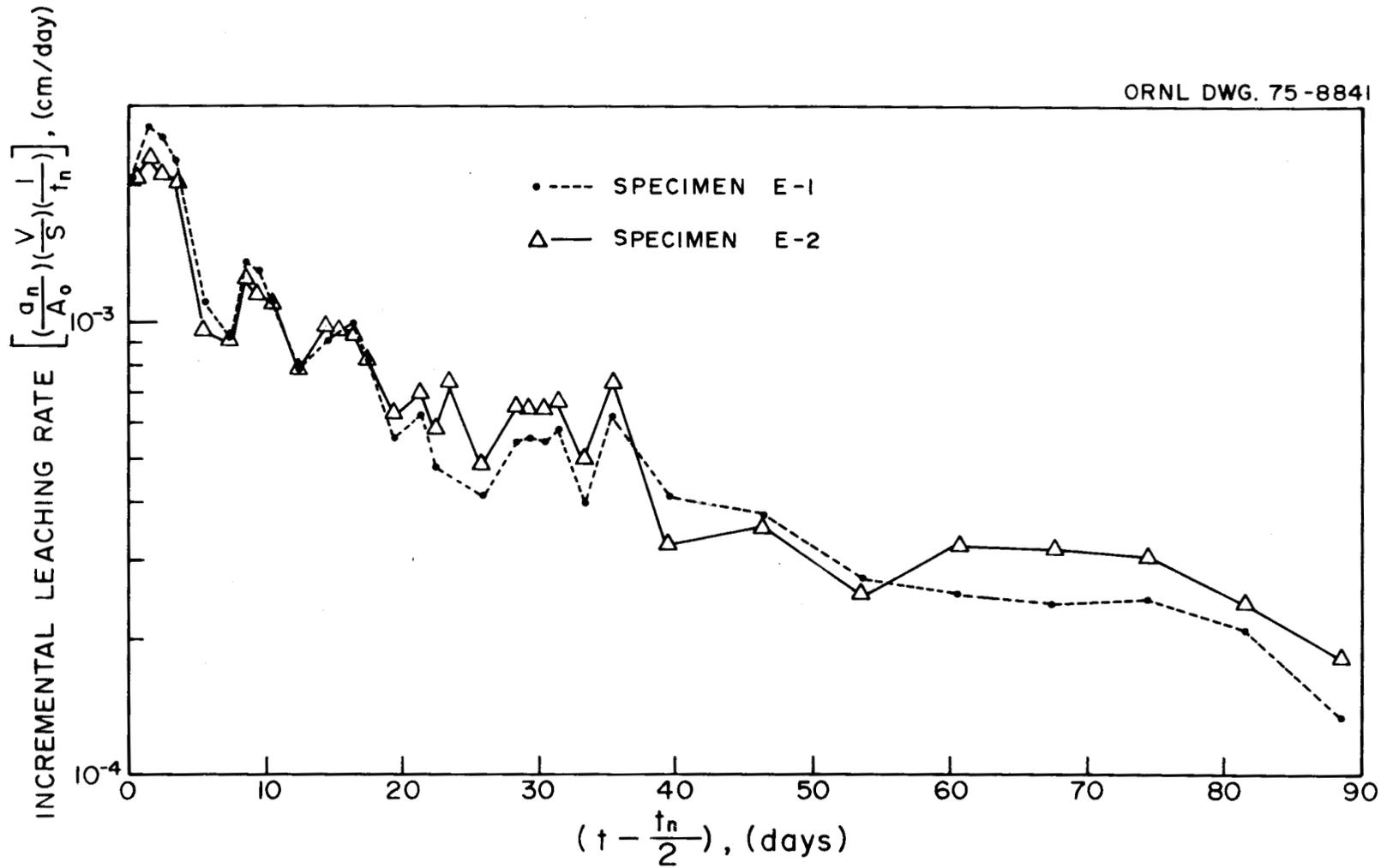


Fig. 5. Incremental leaching rates for Specimens E-1 and E-2, which contain 9.05% iodine as barium iodate plus 0.5% sodium silicate. See Tables A-12 and A-13 for raw data.

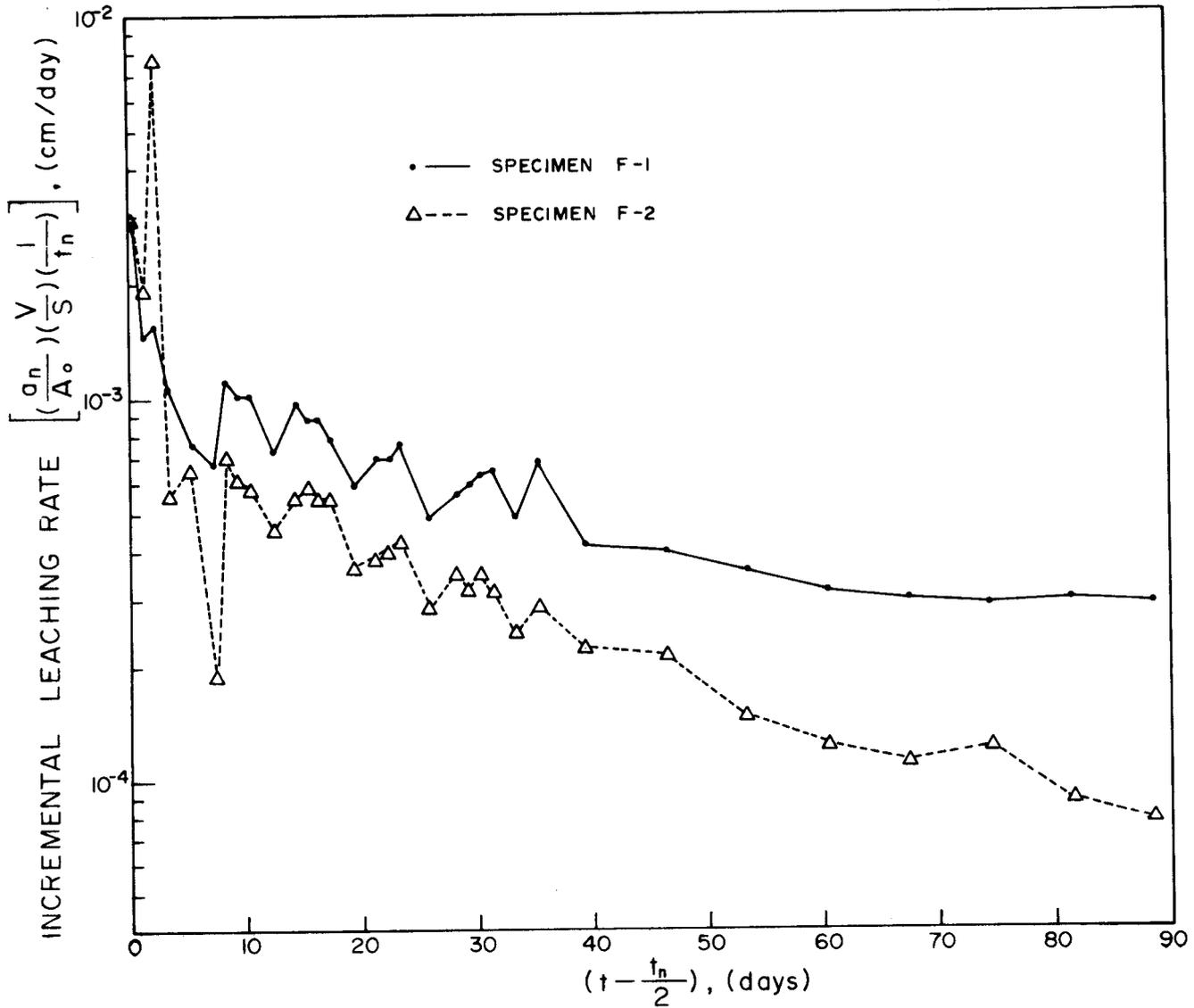


Fig. 6. Incremental leaching rates for Specimens F-1 and F-2, which contain 9.05% iodine as barium iodate plus 0.5% fluosilicic acid. See Tables A-14 and A-15 for raw data.

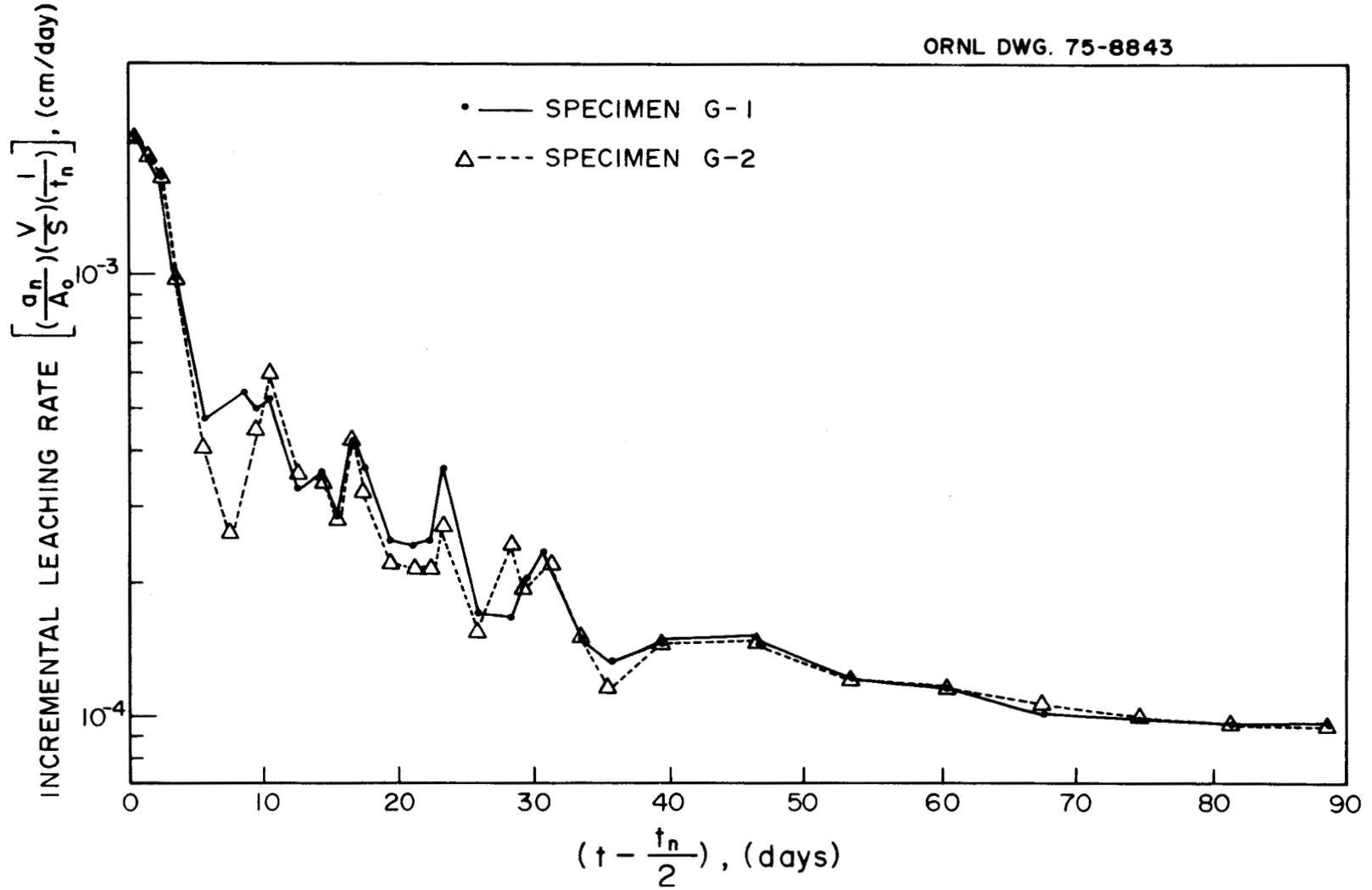


Fig. 7. Incremental leaching rates for Specimens G-1 and G-2, which contain 9.05% iodine as barium iodate plus 0.9% butyl stearate. See Tables A-16 and A-17 for raw data.

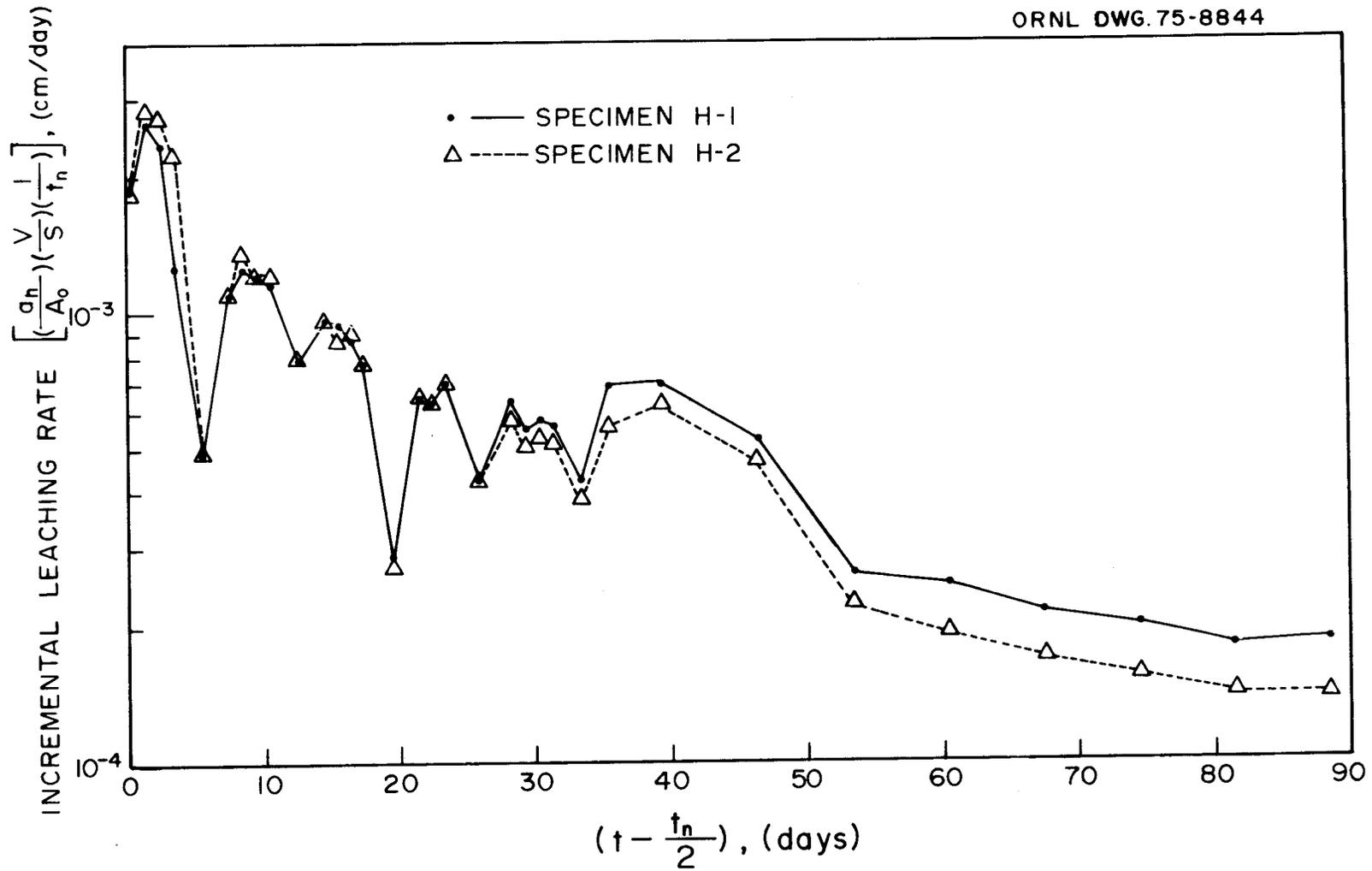


Fig. 8. Incremental leaching rates of Specimens H-1 and H-2, which contain 9.05% iodine as barium iodate plus 1.2% polybutene 24 and 0.5% Aerosol OT. See Tables A-18 and A-19 for raw data.

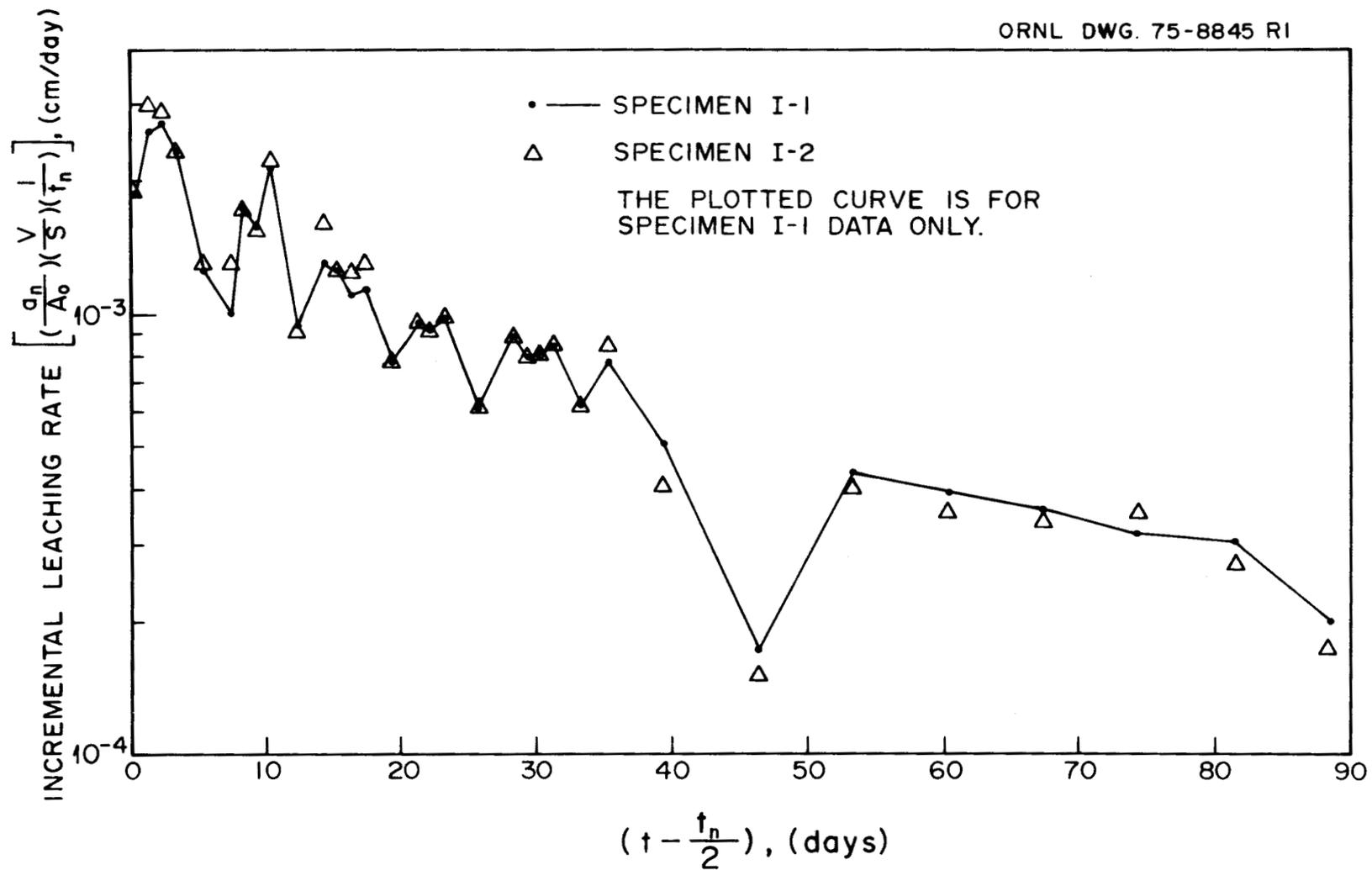


Fig. 9. Incremental leaching rates of Specimens I-1 and I-2, which contain 9.05% iodine as barium iodate plus 1.4% lead monoxide. See Tables A-20 and A-21 for raw data.

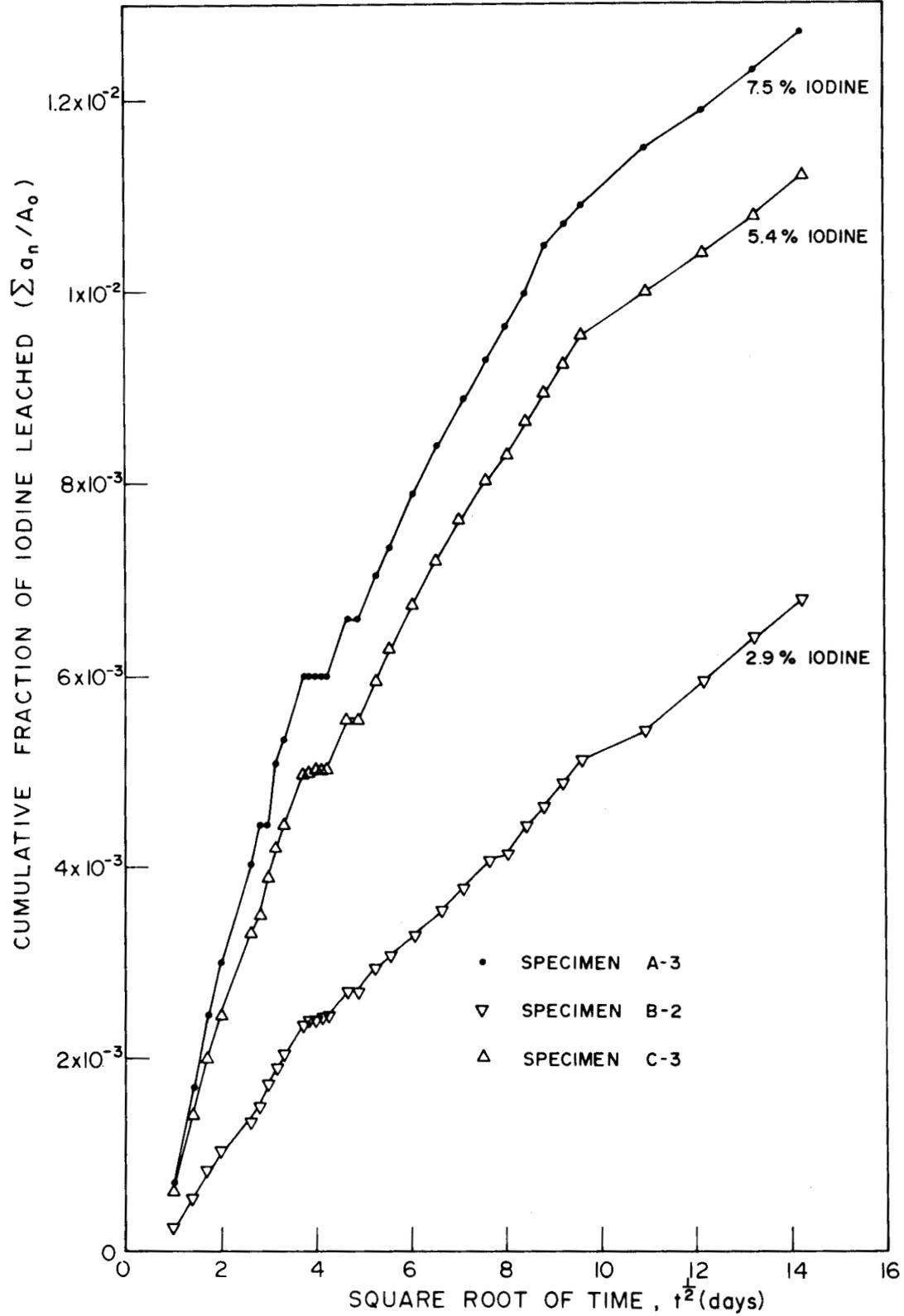


Fig. 10. Effect of iodine content on the leaching rates of Specimens A-3, B-2, and C-3.

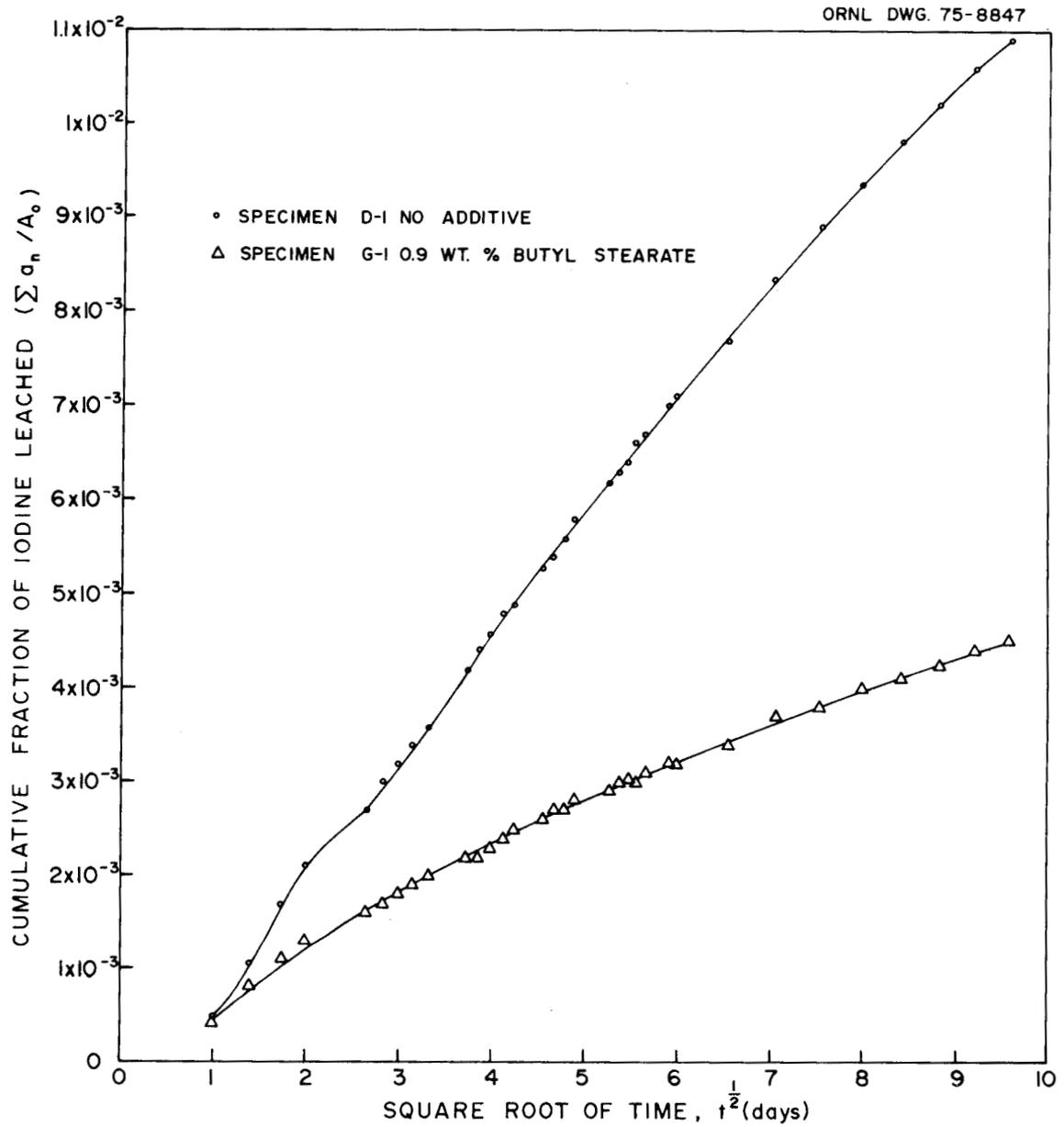


Fig. 11. Effect of 0.9% butyl stearate additive on the leaching rates of specimens which contain 9.05% iodine as barium iodate.

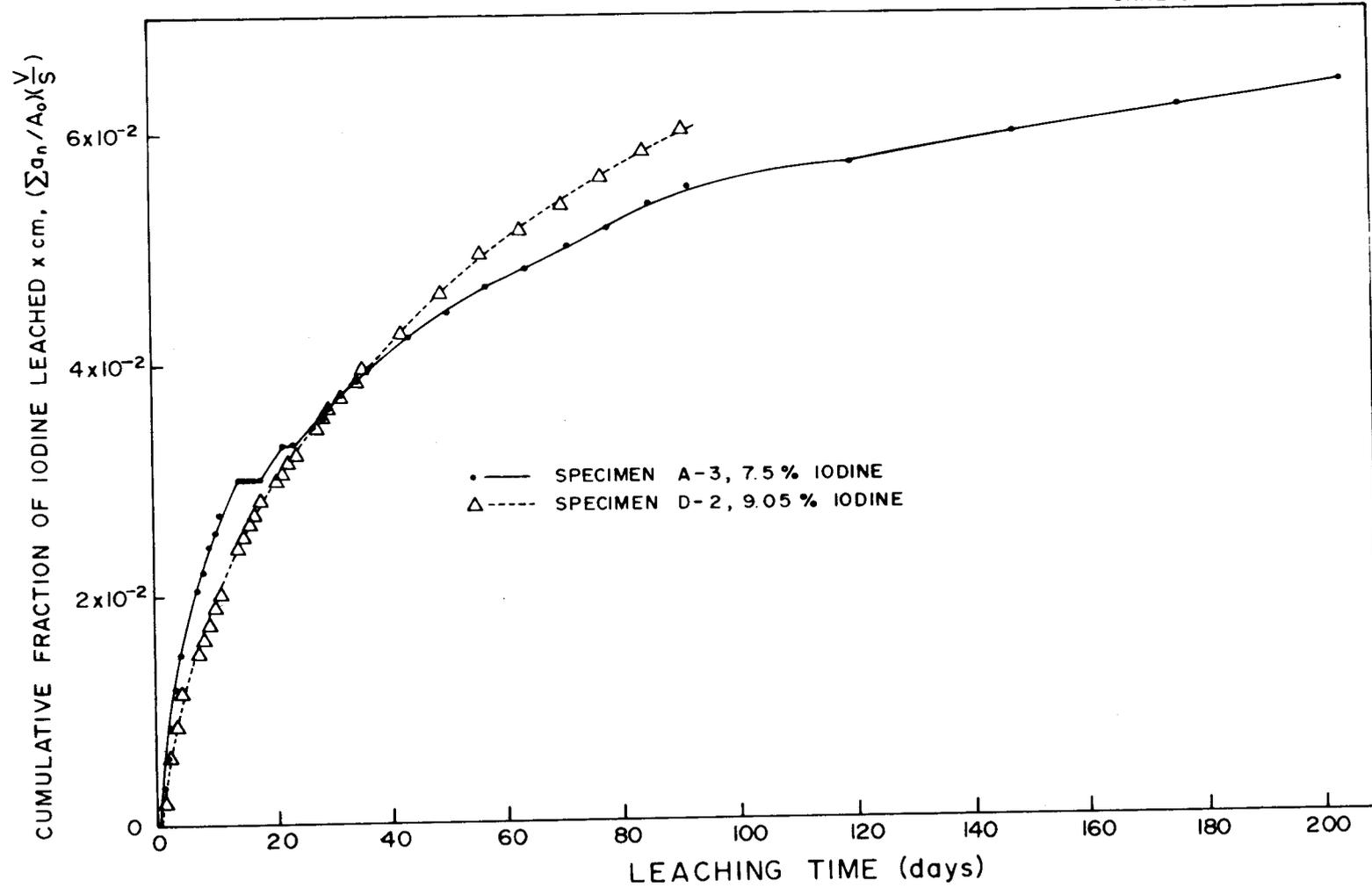
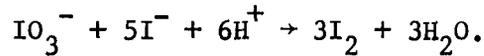
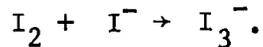


Fig. 12. Diminished effect of varying the iodine content from 7.5% to 9.05% on the leaching rates of Specimens A-3 and D-2. (Compare with results shown in Fig. 10)

The iodate is reduced with iodide ion (KI) in a slightly acid solution, as follows:



In the presence of excess KI, the triiodide ion is formed, as follows:



This ion absorbs strongly at wavelengths of 288 and 352 nm. Molar extinction coefficients are 40,000 and 26,400, respectively.<sup>12</sup>

Although the 288-nm band is slightly more sensitive, we used that at 352 nm because of its greater stability and reproducibility on our instrument, a Beckman DB-G spectrophotometer. With this method we were able to determine the iodine leached from the sample to a lower limit of about 0.01  $\mu\text{g}$  I/ml. Before employing the method, we tested it on solutions that contained known amounts of iodate and had been in contact with crushed concrete for several hours.

Examination of the specimens revealed no signs of cracking, except for occasional shallow surface flaking ("crazing") which seemed to follow no particular pattern with change in composition.

#### 4. RESULTS

The results are presented in the form of incremental leaching plots for each series in Figs. 1-9. In cases where the reproducibility between duplicate or triplicate specimens is good, only a single line is plotted; plots are made for separate specimens where there is significant divergence between specimens. The functions plotted are those recommended by Hespe for the IAEA<sup>11</sup> and by Godbee and Joy,<sup>13</sup> that is,  $(a_n/A_o)(V/S)(1/t_n)$  vs  $(t - t_n/2)$ , where

$a_n$  = weight of iodine leached per leaching period,  $\mu\text{g}$ ,

$A_o$  = the initial iodine content of the specimen,  $\mu\text{g}$ ,

$V$  = volume of the specimen,  $\text{cm}^3$ ,

$S$  = apparent surface area of the specimen,  $\text{cm}^2$ ,

$t$  = total leaching time, days,

$t_n$  = sampling period, days.

In addition, numerical data are presented in Tables A-1 through A-21 in the Appendix. Cumulative leaching data are plotted for certain representative specimens in Figs. 10-12 as a function of leaching time or of the square root of the leaching time.

It should be noted that the units of the incremental ordinate above are centimeters per day. These values can be readily converted to grams per square centimeter per day, if this unit is preferred, by multiplying by the density, mass/volume. The volume for each specimen is  $98.2 \text{ cm}^3$ , while the mass varies between about 178 and 188 g. Taking the mean value of 183 g for the mass gives an effective density of 1.81 for the multiplication factor. The error introduced by using this mean value of the mass rather than that listed in the appropriate table (Tables A-1 through A-21, Appendix) will be of the same order of magnitude as, or lower than, the deviation in leaching rates of duplicate specimens.

The large irregularities seen between the 10th and 25th days of the leach cycle for Series A, B, and C (Figs. 1-3), which were run simultaneously, are too large and agree too closely with each other to have been caused by changes in the sampling period alone. It appears likely that they were the result of constant but undetected errors in the operation of our spectrophotometer. We were, in fact, experiencing difficulty with the instrument during this period, although it was normally checked out on a known solution whenever we were in doubt of its reliability. We have been unable to positively identify the cause of the phenomenon. Operator error, which was considered at the time, seems unlikely since the abnormally low rates continued for several sampling periods. Assuming that such errors did occur, the subsequently reported values for the fraction leached are, of course, lower than they should be. Such errors would not be cumulative for incremental rates. Sudden and unpredictable changes in leaching rates have been observed by others, particularly when irregular leaching periods were employed;<sup>14</sup> however, they are not normally of this size and would not correlate as well between specimens as these do. We plan to repeat leaching tests on at least one of this series of experiments to demonstrate that the abnormal effect seen was indeed caused by some systematic error.

In addition to plots of incremental leaching data, the IAEA<sup>11</sup> also approves of plotting the cumulative fraction of total iodine content leached,  $(a_n/A_0)$ , vs the square root of the leaching time,  $t^{1/2}$ . Use of the square-root function is convenient in that it compresses the abscissa, thus simplifying the graphical presentation of the results of long-term leaching tests.\* Approval by the IAEA of this method of plotting is apparently based on the idea that, since diffusion theory predicts a straight-line plot, one can extrapolate to longer leaching times with greater confidence. In our experiments the leaching process does not appear to be this simple, as shown by the fact that most plots only approach linearity over sections of the curve, that slopes change rather abruptly at times, and that many (probably most) of the "linear" plots do not pass through the origin as predicted by theory (Figs. 10 and 11). A good discussion of the various modes of plotting is given by Godbee and Joy<sup>13</sup> and will not be reviewed here.

Data from representative specimens of Series A, B, and C are presented as square-root plots in Fig. 10. The specimens, A-3 (7.5% iodine), B-2 (2.9% iodine), and C-3 (5.4% iodine) were each selected as being most nearly the median specimens for their particular series. The increase in leaching rate with increase in iodine content is apparent, although the effect diminishes with increasing iodine content. When rates for Specimen D-2 (9.05% iodine, the high specimen of the two runs) are compared with those for A-3, the early parts of the curves overlap well within experimental error. These data are plotted vs time in Fig. 12.\*\* Use of the data from Specimen D-1 (the low member of the D series) would overlap even more.

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\*An even more efficient method of compressing the plots is to use the log of the time as the abscissa. However, the IAEA has not expressed approval of this type of plot.

\*\*This type of plot is included as a demonstration of the difference between the shapes of the plots vs  $t$  and vs  $t^{1/2}$ . It will be noted that the ordinate in Fig. 12 is "normalized" by multiplying the fraction leached by the volume/surface ratio. In Figs. 10 and 11 this normalization is omitted. The V/S for all our samples was constant at 5.0 cm. This makes normalization unnecessary for comparing leach rates of our own specimens.

Specimens were run in which small amounts of the following additives (Table 3) were used in an effort to minimize the loss of iodine by leaching from a concrete containing 9.05 wt % iodine:

0.5%  $\text{Na}_2\text{SiO}_3$  (Series E)

0.5%  $\text{H}_2\text{SiF}_6$  (Series F)

0.9% butyl stearate (Series G)

1.2% polybutene 24 plus 0.5% Aerosol OT (Series H)

1.4% PbO (Series I).

All of these additives except PbO caused some improvement in short-term leach rates over the original composition (Series D). The greatest improvement was observed with the specimens to which butyl stearate was added; leaching rates for these were only about one-half to one-third as high as those of the specimens without any additive (Fig. 11), and the duplication of leach rates between the two leached specimens was good. The beneficial effect of adding butyl stearate is shown graphically in Fig. 11, where cumulative leaching data are plotted vs  $t^{1/2}$  for Specimens D-1 (9.05% iodine, no additive) and G-1 (9.05% iodine, 0.9% butyl stearate). For a leaching time of 50 days, for example, the fraction of iodine leached is only  $3.7 \times 10^{-3}$  for G-1 vs  $8.3 \times 10^{-3}$  for D-1.

Among the other additives, fluosilicic acid,  $\text{H}_2\text{SiF}_6$ , appears to be especially worthy of further investigation, even though there was an exceptionally wide discrepancy between the leach rates of the two specimens tested (Fig. 6).

These data show that the maximum content of iodine incorporated thus far can probably be further increased without a major increase in the relative leaching rate and that further investigation of additives will be worthwhile. At the same time it must be emphasized that, even if leaching curves for two specimens coincide, the total iodine (and hence the total activity) leached will still be higher for specimens with a higher iodine content since the ordinate is expressed in terms of the fraction of iodine leached.

The untreated data from leaching tests on all specimens tested in Series A through I are tabulated in Tables A-1 through A-21 (Appendix).

A 5-metric ton/day LMFBR processing plant will produce 1.605 kg of iodine per day (Table 4).<sup>5</sup> This amount can be incorporated into 0.335 ft<sup>3</sup> of concrete if the concrete contains 9.05 wt % iodine. Decay heat from <sup>129</sup>I would amount to about  $3.3 \times 10^{-3}$  W/metric ton. It will be possible and economical to provide secondary containment (in the form of coatings, metal drums, or both) for these small volumes of solidified waste to ensure that they will not come in contact with water within the foreseeable future. Thus contained, the product, even in its present early stage of development, will be acceptable at any approved repository for radioactive waste.

#### 5. SUMMARY AND CONCLUSIONS

The results obtained from the studies discussed in this report can be summarized as follows:

1. Concrete specimens containing up to 9.05 wt % iodine as barium iodate were cast.
2. The products formed have characteristics which make them acceptable for transportation and for storage at an approved nuclear waste storage facility with minimum containment.
3. Incremental leach rates for the best compositions are below  $1 \times 10^{-4}$  cm/day after 80 days and are still trending downward. Compositions with better leaching characteristics can probably be developed.
4. The maximum iodine content consistent with low leach rates has not been established. Much further work needs to be done to optimize the leach rate and to establish the maximum iodine content consistent with this leach rate.
5. The usefulness of the present work depends on the adoption of a processing scheme (or schemes) for segregating radioactive iodine in the form of iodate.

#### 6. PLANS AND RECOMMENDATIONS FOR FUTURE WORK

The extent of the additional work that needs to be done with regard to the fixation of iodine in solid iodates is heavily dependent upon the acceptance of the Iodox process, or an alternative process, for isolating iodine in the form of iodate. Assuming that Iodox is

Table 4. Fission product iodine from spent LMFBR fuel and its decay

Basis: 1 metric ton of fuel. Calculated by ORIGEN Code.<sup>5</sup>

| Isotope          | Time after discharge from reactor |                   |                       |                       |                       |                       |                        |                        |                       |      |
|------------------|-----------------------------------|-------------------|-----------------------|-----------------------|-----------------------|-----------------------|------------------------|------------------------|-----------------------|------|
|                  | 0 days                            |                   | 90 days               |                       | 150 days              |                       | 365 days               |                        | 10 yrs                |      |
|                  | Ci                                | g                 | Ci                    | g                     | Ci                    | g                     | Ci                     | g                      | Ci                    | g    |
| <sup>127</sup> I | 0                                 | 74.6              | 0                     | 76.8                  | 0                     | 77.5                  | 0                      | 78.6                   | 0                     | 78.9 |
| <sup>129</sup> I | $3.90 \times 10^{-2}$             | 239               | $3.94 \times 10^{-2}$ | 241                   | $3.94 \times 10^{-2}$ | 242                   | $3.94 \times 10^{-2}$  | 242                    | $3.94 \times 10^{-2}$ | 242  |
| <sup>131</sup> I | $1.40 \times 10^6$                | 11.3              | $6.16 \times 10^2$    | $4.97 \times 10^{-3}$ | 3.51                  | $2.84 \times 10^{-5}$ | $3.15 \times 10^{-8}$  | $2.54 \times 10^{-15}$ | 0                     | 0    |
| <sup>132</sup> I | $1.62 \times 10^6$                | Neg. <sup>a</sup> | $7.40 \times 10^{-3}$ | Neg. <sup>a</sup>     | $2.14 \times 10^{-8}$ | Neg. <sup>a</sup>     | $2.50 \times 10^{-28}$ | Neg. <sup>a</sup>      | 0                     | 0    |

<sup>a</sup>Negligible.

accepted, the following are a few of the investigations which should be made (not necessarily in the order of their importance):

1. Evaluate the effect of aging iodate precipitates before their incorporation into concrete.
2. Determine whether there is any appreciable difference in the behavior of  $\text{HIO}_3$  and  $\text{HI}_3\text{O}_8$ .
3. Develop a satisfactory method for producing lead iodate suitable for waste disposal.
4. Evaluate the feasibility of using the iodates of silver or mercury.
5. Develop techniques for producing a more uniform and less leachable final product. This may involve mechanical innovations as well as investigations of additives and combinations of additives.
6. Determine a practical upper limit to the iodine content of the concrete product.

## 7. REFERENCES

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APPENDIX: LEACHING DATA FOR SOLIDS CONTAINING IODINE AS BARIUM IODATE

Table A-1. Leaching data for Specimen A-1 containing 7.5 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 13.5 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 8,100                            | 8,100                       | a                    |
| 1                     | 2                           | 13,260                           | 21,360                      | a                    |
| 1                     | 3                           | 9,540                            | 30,900                      | a                    |
| 1                     | 4                           | 7,380                            | 38,280                      | 11.65                |
| 3                     | 7                           | 13,560                           | 51,840                      | 11.9                 |
| 1                     | 8                           | 5,010                            | 56,850                      | 11.5                 |
| 1                     | 9                           | 4,920                            | 61,770                      | 11.5                 |
| 1                     | 10                          | 4,230                            | 66,000                      | 11.5                 |
| 1                     | 11                          | 3,360                            | 69,360                      | 11.45                |
| 3                     | 14                          | 9,030                            | 78,390                      | 11.8                 |
| 1                     | 15                          | 255                              | 78,645                      | 11.5                 |
| 1                     | 16                          | 180                              | 78,825                      | 11.45                |
| 1                     | 17                          | 169                              | 78,994                      | 11.4                 |
| 1                     | 18                          | 49                               | 79,043                      | 11.4                 |
| 4                     | 22                          | 7,395                            | 86,438                      | 11.7                 |
| 2                     | 24                          | 26                               | 86,464                      | 11.55                |
| 4                     | 28                          | 6,300                            | 92,764                      | 11.6                 |
| 3                     | 31                          | 4,395                            | 97,159                      | 11.6                 |
| 6                     | 37                          | 7,164                            | 104,323                     | 11.9                 |
| 7                     | 44                          | 7,044                            | 111,367                     | 11.75                |
| 7                     | 51                          | 5,961                            | 117,328                     | 11.7                 |
| 7                     | 58                          | 5,235                            | 122,563                     | 11.6                 |
| 7                     | 65                          | 3,564                            | 126,127                     | 11.6                 |
| 7                     | 72                          | 4,566                            | 130,693                     | 11.5                 |
| 7                     | 79                          | 4,161                            | 134,854                     | 11.5                 |
| 7                     | 86                          | 4,035                            | 138,889                     | 11.4                 |
| 7                     | 93                          | 3,780                            | 142,669                     | 11.45                |
| 28                    | 121                         | 6,660                            | 149,329                     | 11.50                |
| 28                    | 149                         | 5,820                            | 155,149                     | 10.7                 |
| 28                    | 177                         | 5,640                            | 160,739                     | 11.4                 |
| 28                    | 205                         | 5,160                            | 165,899                     | 10.75                |

<sup>a</sup>Not determined.

Table A-2. Leaching data for Specimen A-2 containing 7.5 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 13.5 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|---------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$               | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 10,020                          | 10,020                      | a                    |
| 1                     | 2                           | 14,280                          | 24,300                      | a                    |
| 1                     | 3                           | 10,380                          | 34,680                      | a                    |
| 1                     | 4                           | 8,040                           | 42,720                      | 11.7                 |
| 3                     | 7                           | 14,820                          | 57,540                      | 11.9                 |
| 1                     | 8                           | 5,190                           | 62,730                      | 11.5                 |
| 1                     | 9                           | 5,250                           | 67,980                      | 11.5                 |
| 1                     | 10                          | 4,530                           | 72,510                      | 11.5                 |
| 1                     | 11                          | 4,200                           | 76,710                      | 11.45                |
| 3                     | 14                          | 9,630                           | 86,340                      | 11.8                 |
| 1                     | 15                          | 264                             | 86,604                      | 11.5                 |
| 1                     | 16                          | 185                             | 86,789                      | 11.45                |
| 1                     | 17                          | 180                             | 86,969                      | 11.4                 |
| 1                     | 18                          | 49                              | 87,018                      | 11.4                 |
| 4                     | 22                          | 8,166                           | 95,184                      | 11.7                 |
| 2                     | 24                          | 32                              | 95,216                      | 11.55                |
| 4                     | 28                          | 6,612                           | 101,828                     | 11.5                 |
| 3                     | 31                          | 4,806                           | 106,634                     | 11.6                 |
| 6                     | 37                          | 7,575                           | 114,209                     | 11.9                 |
| 7                     | 44                          | 7,038                           | 121,247                     | 11.7                 |
| 7                     | 51                          | 6,804                           | 128,051                     | 11.7                 |
| 7                     | 58                          | 6,180                           | 134,231                     | 11.6                 |
| 7                     | 65                          | 4,320                           | 138,551                     | 11.6                 |
| 7                     | 72                          | 5,226                           | 143,777                     | 11.5                 |
| 7                     | 79                          | 4,680                           | 148,457                     | 11.45                |
| 7                     | 86                          | 4,404                           | 152,861                     | 11.45                |
| 7                     | 93                          | 3,960                           | 156,821                     | 11.45                |
| 28                    | 121                         | 6,660                           | 163,481                     | 11.5                 |
| 28                    | 149                         | 6,600                           | 170,081                     | 10.7                 |
| 28                    | 177                         | 5,526                           | 175,607                     | 11.4                 |
| 28                    | 205                         | 5,340                           | 180,947                     | 10.75                |

<sup>a</sup>Not determined.

Table A-3. Leaching data for Specimen A-3 containing 7.5 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 13.5 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$                | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 9,480                            | 9,480                       | a                    |
| 1                     | 2                           | 13,260                           | 22,740                      | a                    |
| 1                     | 3                           | 10,320                           | 33,060                      | a                    |
| 1                     | 4                           | 7,800                            | 40,860                      | 11.7                 |
| 3                     | 7                           | 14,040                           | 54,900                      | 12.0                 |
| 1                     | 8                           | 4,860                            | 59,760                      | 11.5                 |
| 1                     | 9                           | 4,680                            | 64,440                      | 11.5                 |
| 1                     | 10                          | 4,140                            | 68,580                      | 11.5                 |
| 1                     | 11                          | 3,690                            | 72,270                      | 11.45                |
| 3                     | 14                          | 8,430                            | 80,700                      | 11.6                 |
| 1                     | 15                          | 246                              | 80,946                      | 11.5                 |
| 1                     | 16                          | 163                              | 81,109                      | 11.45                |
| 1                     | 17                          | 170                              | 81,279                      | 11.45                |
| 1                     | 18                          | 56                               | 81,335                      | 11.45                |
| 4                     | 22                          | 7,680                            | 89,015                      | 11.7                 |
| 2                     | 24                          | 24                               | 89,039                      | 11.6                 |
| 4                     | 28                          | 6,126                            | 95,165                      | 11.5                 |
| 3                     | 31                          | 4,515                            | 99,680                      | 11.6                 |
| 6                     | 37                          | 7,164                            | 106,844                     | 11.9                 |
| 7                     | 44                          | 6,849                            | 113,693                     | 11.7                 |
| 7                     | 51                          | 6,309                            | 120,002                     | 11.7                 |
| 7                     | 58                          | 5,964                            | 125,966                     | 11.65                |
| 7                     | 65                          | 4,164                            | 130,130                     | 11.55                |
| 7                     | 72                          | 5,235                            | 135,365                     | 11.5                 |
| 7                     | 79                          | 4,389                            | 139,754                     | 11.5                 |
| 7                     | 86                          | 4,386                            | 144,140                     | 11.45                |
| 7                     | 93                          | 3,735                            | 147,875                     | 11.45                |
| 28                    | 121                         | 6,660                            | 154,535                     | 11.5                 |
| 28                    | 149                         | 6,138                            | 160,673                     | 10.7                 |
| 28                    | 177                         | 5,358                            | 166,031                     | 11.4                 |
| 28                    | 205                         | 5,520                            | 171,551                     | 10.75                |

<sup>a</sup>Not determined.

Table A-4. Leaching data for Specimen B-1 containing 2.9 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 5.22 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$                | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 1,140                            | 1,140                       | a                    |
| 1                     | 2                           | 1,140                            | 2,280                       | a                    |
| 1                     | 3                           | 1,140                            | 3,420                       | a                    |
| 1                     | 4                           | 1,080                            | 4,500                       | 11.7                 |
| 3                     | 7                           | 1,260                            | 5,760                       | 11.75                |
| 1                     | 8                           | 870                              | 6,630                       | 11.5                 |
| 1                     | 9                           | 930                              | 7,560                       | 11.45                |
| 1                     | 10                          | 660                              | 8,220                       | 11.3                 |
| 1                     | 11                          | 600                              | 8,820                       | 11.25                |
| 3                     | 14                          | 1,215                            | 10,035                      | 11.8                 |
| 1                     | 15                          | 63                               | 10,098                      | 11.45                |
| 1                     | 16                          | 53                               | 10,151                      | 11.2                 |
| 1                     | 17                          | 48                               | 10,199                      | 11.2                 |
| 1                     | 18                          | 3                                | 10,202                      | 11.25                |
| 4                     | 22                          | 1,089                            | 11,291                      | 11.6                 |
| 2                     | 24                          | 7                                | 11,298                      | 11.55                |
| 4                     | 28                          | 1,026                            | 12,324                      | 11.55                |
| 3                     | 31                          | 804                              | 13,128                      | 11.45                |
| 6                     | 37                          | 975                              | 14,103                      | 11.7                 |
| 7                     | 44                          | 1,020                            | 15,123                      | 11.6                 |
| 7                     | 51                          | 1,095                            | 16,218                      | 11.5                 |
| 7                     | 58                          | 1,080                            | 17,298                      | 11.5                 |
| 7                     | 65                          | 292                              | 17,590                      | 11.55                |
| 7                     | 72                          | 1,170                            | 18,760                      | 11.4                 |
| 7                     | 79                          | 1,080                            | 19,840                      | 11.4                 |
| 7                     | 86                          | 1,260                            | 21,100                      | 11.45                |
| 7                     | 93                          | 1,293                            | 22,393                      | 11.5                 |
| 28                    | 121                         | 1,344                            | 23,737                      | 11.6                 |
| 28                    | 149                         | 1,632                            | 25,369                      | 10.0                 |
| 28                    | 177                         | 2,946                            | 28,315                      | 11.4                 |
| 28                    | 205                         | 1,710                            | 30,025                      | 10.7                 |

<sup>a</sup>Not determined.

Table A-5. Leaching data for Specimen B-2 containing 2.9 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 5.22 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$                | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1 ]                         | 1,410                            | 1,410                       | a                    |
| 1                     | 2                           | 1,560                            | 2,970                       | a                    |
| 1                     | 3                           | 1,380                            | 4,350                       | a                    |
| 1                     | 4                           | 1,080                            | 5,430                       | 11.6                 |
| 3                     | 7                           | 1,560                            | 6,990                       | 11.95                |
| 1                     | 8                           | 960                              | 7,950                       | 11.45                |
| 1                     | 9                           | 1,200                            | 9,150                       | 11.5                 |
| 1                     | 10                          | 840                              | 9,990                       | 11.4                 |
| 1                     | 11                          | 750                              | 10,740                      | 11.35                |
| 3                     | 14                          | 1,650                            | 12,390                      | 11.8                 |
| 1                     | 15                          | 69                               | 12,459                      | 11.45                |
| 1                     | 16                          | 69                               | 12,528                      | 11.25                |
| 1                     | 17                          | 62                               | 12,590                      | 11.2                 |
| 1                     | 18                          | 15                               | 12,605                      | 11.2                 |
| 4                     | 22                          | 1,275                            | 13,880                      | 11.65                |
| 2                     | 24                          | 14                               | 13,894                      | 11.6                 |
| 4                     | 28                          | 1,200                            | 15,094                      | 11.5                 |
| 3                     | 31                          | 984                              | 16,078                      | 11.5                 |
| 6                     | 37                          | 1,155                            | 17,233                      | 11.75                |
| 7                     | 44                          | 1,269                            | 18,502                      | 11.65                |
| 7                     | 51                          | 1,380                            | 19,882                      | 11.5                 |
| 7                     | 58                          | 1,371                            | 21,253                      | 11.5                 |
| 7                     | 64                          | 460                              | 21,713                      | 11.5                 |
| 7                     | 72                          | 1,320                            | 23,033                      | 11.3                 |
| 7                     | 79                          | 1,203                            | 24,236                      | 11.3                 |
| 7                     | 86                          | 1,260                            | 25,496                      | 11.35                |
| 7                     | 93                          | 1,293                            | 26,789                      | 11.35                |
| 28                    | 121                         | 1,458                            | 28,247                      | 11.55                |
| 28                    | 149                         | 2,535                            | 30,782                      | 10.0                 |
| 28                    | 177                         | 3,042                            | 33,824                      | 11.4                 |
| 28                    | 205                         | 1,719                            | 35,543                      | 10.7                 |

<sup>a</sup>Not determined.

Table A-6. Leaching data for Specimen B-3 containing 2.9 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 5.22 g

Specimen  
 Mass (M) = 180 g  
 Volume (M) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$                | Cumulative ( $\Sigma t_n$ ) |                      |
| 1                     | 1                           | 1,800                            | 1,800                       | a                    |
| 1                     | 2                           | 1,740                            | 3,540                       | a                    |
| 1                     | 3                           | 1,260                            | 4,800                       | a                    |
| 1                     | 4                           | 1,020                            | 5,820                       | 11.65                |
| 3                     | 7                           | 1,620                            | 7,440                       | 11.95                |
| 1                     | 8                           | 960                              | 8,400                       | 11.45                |
| 1                     | 9                           | 1,140                            | 9,540                       | 11.5                 |
| 1                     | 10                          | 750                              | 10,290                      | 11.45                |
| 1                     | 11                          | 750                              | 11,040                      | 11.35                |
| 3                     | 14                          | 1,590                            | 12,630                      | 11.8                 |
| 1                     | 15                          | 60                               | 12,690                      | 11.45                |
| 1                     | 16                          | 61                               | 12,751                      | 11.2                 |
| 1                     | 17                          | 68                               | 12,819                      | 11.2                 |
| 1                     | 18                          | 15                               | 12,834                      | 11.3                 |
| 4                     | 22                          | 1,338                            | 14,172                      | 11.6                 |
| 2                     | 24                          | 32                               | 14,204                      | 11.6                 |
| 4                     | 28                          | 1,266                            | 15,470                      | 11.5                 |
| 3                     | 31                          | 900                              | 16,370                      | 11.45                |
| 6                     | 37                          | 1,269                            | 17,639                      | 11.7                 |
| 7                     | 44                          | 1,263                            | 18,902                      | 11.6                 |
| 7                     | 51                          | 1,380                            | 20,282                      | 11.5                 |
| 7                     | 58                          | 1,350                            | 21,632                      | 11.5                 |
| 7                     | 65                          | 513                              | 22,145                      | 11.5                 |
| 7                     | 72                          | 1,290                            | 23,435                      | 11.4                 |
| 7                     | 79                          | 1,203                            | 24,638                      | 11.3                 |
| 8                     | 86                          | 1,260                            | 25,898                      | 11.3                 |
| 7                     | 93                          | 1,293                            | 27,191                      | 11.35                |
| 28                    | 121                         | 1,446                            | 28,637                      | 11.55                |
| 28                    | 149                         | 1,740                            | 30,377                      | 10.0                 |
| 28                    | 177                         | 3,216                            | 33,593                      | 11.4                 |
| 28                    | 205                         | 1,863                            | 35,456                      | 10.7                 |

<sup>a</sup>Not determined.

Table A-7. Leaching data for Specimen C-1 containing 5.4 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 9.72 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental $a_n$                | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,450                            | 6,450                       | a                    |
| 1                     | 2                           | 7,860                            | 14,310                      | a                    |
| 1                     | 3                           | 5,580                            | 19,890                      | a                    |
| 1                     | 4                           | 4,380                            | 24,270                      | 11.65                |
| 3                     | 7                           | 8,100                            | 32,370                      | 11.95                |
| 1                     | 8                           | 3,060                            | 35,430                      | 11.5                 |
| 1                     | 9                           | 3,000                            | 38,430                      | 11.5                 |
| 1                     | 10                          | 2,550                            | 40,980                      | 11.5                 |
| 1                     | 11                          | 2,400                            | 43,380                      | 11.45                |
| 3                     | 14                          | 5,880                            | 49,260                      | 11.8                 |
| 1                     | 15                          | 180                              | 49,440                      | 11.45                |
| 1                     | 16                          | 146                              | 49,586                      | 11.4                 |
| 1                     | 17                          | 139                              | 49,725                      | 11.4                 |
| 1                     | 18                          | 38                               | 49,763                      | 11.45                |
| 4                     | 22                          | 5,235                            | 54,998                      | 11.75                |
| 2                     | 24                          | 30                               | 55,028                      | 11.6                 |
| 4                     | 28                          | 4,500                            | 59,528                      | 11.5                 |
| 3                     | 31                          | 3,240                            | 62,768                      | 11.65                |
| 6                     | 37                          | 5,235                            | 68,003                      | 11.95                |
| 7                     | 44                          | 4,995                            | 72,998                      | 11.8                 |
| 7                     | 51                          | 4,939                            | 77,937                      | 11.7                 |
| 7                     | 58                          | 4,620                            | 82,557                      | 11.7                 |
| 7                     | 65                          | 2,649                            | 85,206                      | 11.5                 |
| 7                     | 72                          | 3,906                            | 89,112                      | 11.5                 |
| 7                     | 79                          | 3,435                            | 92,547                      | 11.45                |
| 7                     | 86                          | 3,420                            | 95,967                      | 11.4                 |
| 7                     | 93                          | 2,946                            | 98,913                      | 11.45                |
| 28                    | 121                         | 4,860                            | 103,773                     | 11.65                |
| 28                    | 149                         | 4,680                            | 108,453                     | 10.5                 |
| 28                    | 177                         | 4,395                            | 112,848                     | 11.4                 |
| 28                    | 205                         | 3,975                            | 116,823                     | 10.65                |

<sup>a</sup>Not determined.

Table A-8. Leaching data for Specimen C-2 containing 5.4 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 9.72 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 5,310                            | 5,310                       | a                    |
| 1                     | 2                           | 6,360                            | 11,670                      | a                    |
| 1                     | 3                           | 5,100                            | 16,770                      | a                    |
| 1                     | 4                           | 5,940                            | 22,710                      | 11.65                |
| 3                     | 7                           | 7,380                            | 30,090                      | 11.9                 |
| 1                     | 8                           | 2,970                            | 33,060                      | 11.5                 |
| 1                     | 9                           | 2,850                            | 35,910                      | 11.5                 |
| 1                     | 10                          | 2,520                            | 38,430                      | 11.5                 |
| 1                     | 11                          | 2,220                            | 40,650                      | 11.45                |
| 3                     | 14                          | 5,400                            | 46,050                      | 11.8                 |
| 1                     | 15                          | 150                              | 46,200                      | 11.45                |
| 1                     | 16                          | 139                              | 46,339                      | 11.4                 |
| 1                     | 17                          | 134                              | 46,473                      | 11.4                 |
| 1                     | 18                          | 37                               | 46,510                      | 11.4                 |
| 4                     | 22                          | 4,764                            | 51,274                      | 11.7                 |
| 2                     | 24                          | 31                               | 51,305                      | 11.55                |
| 4                     | 28                          | 4,140                            | 55,445                      | 11.5                 |
| 3                     | 31                          | 2,901                            | 58,346                      | 11.6                 |
| 6                     | 37                          | 4,518                            | 62,864                      | 11.95                |
| 7                     | 44                          | 4,635                            | 67,499                      | 11.8                 |
| 7                     | 51                          | 4,500                            | 71,999                      | 11.7                 |
| 7                     | 58                          | 4,275                            | 76,274                      | 11.7                 |
| 7                     | 65                          | 2,145                            | 78,419                      | 11.55                |
| 7                     | 72                          | 3,720                            | 82,139                      | 11.5                 |
| 7                     | 79                          | 3,210                            | 85,349                      | 11.45                |
| 7                     | 86                          | 3,120                            | 88,469                      | 11.4                 |
| 7                     | 93                          | 3,000                            | 91,469                      | 11.45                |
| 28                    | 121                         | 4,569                            | 96,038                      | 11.65                |
| 28                    | 149                         | 4,800                            | 100,838                     | 10.5                 |
| 28                    | 177                         | 4,386                            | 105,224                     | 11.45                |
| 28                    | 205                         | 4,320                            | 109,544                     | 10.65                |

<sup>a</sup>Not determined.

Table A-9. Leaching data for Specimen C-3 containing 5.4 wt % iodine as barium iodate

Curing time = 77 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 9.72 g

Specimen  
 Mass (M) = 180 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,270                            | 6,270                       | a                    |
| 1                     | 2                           | 7,560                            | 13,830                      | a                    |
| 1                     | 3                           | 5,340                            | 19,170                      | a                    |
| 1                     | 4                           | 4,500                            | 23,670                      | 11.65                |
| 3                     | 7                           | 8,220                            | 31,890                      | 11.9                 |
| 1                     | 8                           | 3,060                            | 34,950                      | 11.5                 |
| 1                     | 9                           | 3,180                            | 38,130                      | 11.6                 |
| 1                     | 10                          | 2,580                            | 40,710                      | 11.5                 |
| 1                     | 11                          | 2,370                            | 43,080                      | 11.45                |
| 3                     | 14                          | 5,730                            | 48,810                      | 11.8                 |
| 1                     | 15                          | 180                              | 48,990                      | 11.5                 |
| 1                     | 16                          | 145                              | 49,135                      | 11.4                 |
| 1                     | 17                          | 138                              | 49,273                      | 11.4                 |
| 1                     | 18                          | 37                               | 49,310                      | 11.4                 |
| 4                     | 22                          | 4,881                            | 54,191                      | 11.75                |
| 2                     | 24                          | 32                               | 54,223                      | 11.55                |
| 4                     | 28                          | 3,855                            | 58,078                      | 11.55                |
| 3                     | 31                          | 3,081                            | 61,159                      | 11.6                 |
| 6                     | 37                          | 4,524                            | 65,683                      | 11.9                 |
| 7                     | 44                          | 4,449                            | 70,132                      | 11.7                 |
| 7                     | 51                          | 4,401                            | 74,533                      | 11.75                |
| 7                     | 58                          | 4,080                            | 78,613                      | 11.7                 |
| 7                     | 65                          | 2,304                            | 80,917                      | 11.55                |
| 7                     | 72                          | 3,390                            | 84,307                      | 11.5                 |
| 7                     | 79                          | 3,090                            | 87,397                      | 11.45                |
| 7                     | 86                          | 2,829                            | 90,226                      | 11.45                |
| 7                     | 93                          | 2,958                            | 93,184                      | 11.45                |
| 28                    | 121                         | 4,200                            | 97,384                      | 11.7                 |
| 28                    | 149                         | 4,284                            | 101,668                     | 10.55                |
| 28                    | 177                         | 3,780                            | 105,448                     | 11.45                |
| 28                    | 205                         | 3,852                            | 109,300                     | 10.65                |

<sup>a</sup>Not determined.

Table A-10. Leaching data for Specimen D-1 containing 9.05 wt % iodine as barium iodate

Curing time = 109 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 7,800                            | 7,800                       | 11.0                 |
| 1                     | 2                           | 9,720                            | 17,520                      | 10.6                 |
| 1                     | 3                           | 10,470                           | 27,990                      | 11.1                 |
| 1                     | 4                           | 6,480                            | 34,470                      | 10.0                 |
| 3                     | 7                           | 11,316                           | 45,786                      | 10.6                 |
| 1                     | 8                           | 3,528                            | 49,314                      | 10.5                 |
| 1                     | 9                           | 4,401                            | 53,715                      | 10.65                |
| 1                     | 10                          | 3,984                            | 57,699                      | 10.1                 |
| 1                     | 11                          | 3,624                            | 61,323                      | 10.6                 |
| 3                     | 14                          | 8,952                            | 70,275                      | 11.5                 |
| 1                     | 15                          | 3,312                            | 73,587                      | 10.6                 |
| 1                     | 16                          | 2,835                            | 76,422                      | 11.2                 |
| 1                     | 17                          | 3,069                            | 79,491                      | 11.1                 |
| 1                     | 18                          | 2,763                            | 82,254                      | 10.5                 |
| 3                     | 21                          | 6,315                            | 88,569                      | 11.5                 |
| 1                     | 22                          | 2,493                            | 91,062                      | 10.1                 |
| 1                     | 23                          | 2,646                            | 93,708                      | 10.2                 |
| 1                     | 24                          | 2,766                            | 96,474                      | 11.2                 |
| 4                     | 28                          | 6,858                            | 103,332                     | 11.7                 |
| 1                     | 29                          | 2,370                            | 105,702                     | 11.2                 |
| 1                     | 30                          | 2,145                            | 107,847                     | 11.15                |
| 1                     | 31                          | 2,235                            | 110,082                     | 10.3                 |
| 1                     | 32                          | 2,262                            | 112,344                     | 11.2                 |
| 3                     | 35                          | 4,938                            | 117,282                     | 11.0                 |
| 1                     | 36                          | 2,520                            | 119,802                     | 10.85                |
| 7                     | 43                          | 9,768                            | 129,570                     | 11.6                 |
| 7                     | 50                          | 10,224                           | 139,794                     | 10.75                |
| 7                     | 57                          | 8,448                            | 148,242                     | 11.0                 |
| 7                     | 64                          | 7,800                            | 156,042                     | 11.0                 |
| 7                     | 71                          | 7,230                            | 163,272                     | 10.9                 |
| 7                     | 78                          | 7,200                            | 170,472                     | 11.25                |
| 7                     | 85                          | 6,720                            | 177,192                     | 11.45                |
| 7                     | 92                          | 5,682                            | 182,874                     | 11.4                 |

Table A-11. Leaching data for Specimen D-2 containing 9.05 wt % iodine as barium iodate

Curing time = 109 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| <u>Leaching time (days)</u>     |                                       | <u>Iodine leached (µg)</u>      |                                       | Final pH of leachate |
|---------------------------------|---------------------------------------|---------------------------------|---------------------------------------|----------------------|
| <u>Incremental</u><br>( $t_n$ ) | <u>Cumulative</u><br>( $\Sigma t_n$ ) | <u>Incremental</u><br>( $a_n$ ) | <u>Cumulative</u><br>( $\Sigma a_n$ ) |                      |
| 1                               | 1                                     | 7,935                           | 7,935                                 | 11.1                 |
| 1                               | 2                                     | 12,720                          | 20,655                                | 10.5                 |
| 1                               | 3                                     | 7,470                           | 28,125                                | 11.0                 |
| 1                               | 4                                     | 9,780                           | 37,905                                | 10.5                 |
| 3                               | 7                                     | 12,000                          | 49,905                                | 10.6                 |
| 1                               | 8                                     | 4,230                           | 54,135                                | 10.7                 |
| 1                               | 9                                     | 5,100                           | 59,235                                | 10.6                 |
| 1                               | 10                                    | 4,875                           | 64,110                                | 10.2                 |
| 1                               | 11                                    | 4,266                           | 68,376                                | 10.7                 |
| 3                               | 14                                    | 11,460                          | 79,836                                | 11.6                 |
| 1                               | 15                                    | 3,666                           | 83,502                                | 10.6                 |
| 1                               | 16                                    | 3,234                           | 86,736                                | 11.2                 |
| 1                               | 17                                    | 3,330                           | 90,066                                | 11.1                 |
| 1                               | 18                                    | 3,093                           | 93,159                                | 10.5                 |
| 3                               | 21                                    | 6,900                           | 100,059                               | 11.5                 |
| 1                               | 22                                    | 2,760                           | 102,819                               | 10.1                 |
| 1                               | 23                                    | 2,880                           | 105,699                               | 10.0                 |
| 1                               | 24                                    | 2,745                           | 108,444                               | 11.2                 |
| 4                               | 28                                    | 7,335                           | 115,779                               | 11.7                 |
| 1                               | 29                                    | 2,550                           | 118,329                               | 11.2                 |
| 1                               | 30                                    | 2,598                           | 120,927                               | 11.1                 |
| 1                               | 31                                    | 2,598                           | 123,525                               | 10.3                 |
| 1                               | 32                                    | 2,400                           | 125,925                               | 11.2                 |
| 3                               | 35                                    | 5,661                           | 131,586                               | 11.0                 |
| 1                               | 36                                    | 2,736                           | 134,322                               | 10.8                 |
| 7                               | 43                                    | 10,440                          | 144,762                               | 11.6                 |
| 7                               | 50                                    | 12,240                          | 157,002                               | 10.75                |
| 7                               | 57                                    | 9,048                           | 166,050                               | 11.0                 |
| 7                               | 64                                    | 8,070                           | 174,120                               | 11.0                 |
| 7                               | 71                                    | 7,968                           | 182,088                               | 10.9                 |
| 7                               | 78                                    | 7,608                           | 189,696                               | 11.25                |
| 7                               | 85                                    | 7,104                           | 196,800                               | 11.45                |
| 7                               | 92                                    | 4,464                           | 201,264                               | 11.4                 |

Table A-12. Leaching data for Specimen E-1 containing 9.05 wt % iodine as barium iodate plus 0.5% sodium silicate

Curing time = 100 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|---------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )           | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,900                           | 6,900                       | 11.2                 |
| 1                     | 2                           | 9,000                           | 15,900                      | 10.3                 |
| 1                     | 3                           | 8,538                           | 24,438                      | 11.1                 |
| 1                     | 4                           | 7,500                           | 31,938                      | 10.0                 |
| 3                     | 7                           | 10,830                          | 42,768                      | 10.4                 |
| 1                     | 8                           | 3,012                           | 45,780                      | 10.4                 |
| 1                     | 9                           | 4,460                           | 50,240                      | 10.6                 |
| 1                     | 10                          | 4,326                           | 54,556                      | 9.9                  |
| 1                     | 11                          | 3,900                           | 58,466                      | 10.6                 |
| 3                     | 14                          | 7,920                           | 66,386                      | 11.2                 |
| 1                     | 15                          | 3,081                           | 69,467                      | 10.8                 |
| 1                     | 16                          | 3,180                           | 72,647                      | 11.2                 |
| 1                     | 17                          | 3,393                           | 76,040                      | 11.1                 |
| 1                     | 18                          | 2,763                           | 78,803                      | 10.3                 |
| 3                     | 21                          | 5,526                           | 84,329                      | 11.35                |
| 1                     | 22                          | 2,100                           | 86,429                      | 10.5                 |
| 1                     | 23                          | 1,623                           | 88,052                      | 10.2                 |
| 1                     | 24                          | 2,343                           | 90,395                      | 11.0                 |
| 4                     | 28                          | 5,484                           | 95,879                      | 11.6                 |
| 1                     | 29                          | 1,842                           | 97,721                      | 11.1                 |
| 1                     | 30                          | 1,848                           | 99,569                      | 11.15                |
| 1                     | 31                          | 1,827                           | 101,396                     | 10.2                 |
| 1                     | 32                          | 1,926                           | 103,322                     | 11.1                 |
| 3                     | 35                          | 3,924                           | 107,246                     | 10.9                 |
| 1                     | 36                          | 2,118                           | 109,364                     | 10.9                 |
| 7                     | 43                          | 9,630                           | 118,994                     | 11.5                 |
| 7                     | 50                          | 8,808                           | 127,802                     | 10.6                 |
| 7                     | 57                          | 6,276                           | 134,078                     | 11.0                 |
| 7                     | 64                          | 5,880                           | 139,958                     | 11.1                 |
| 7                     | 71                          | 5,640                           | 145,598                     | 11.0                 |
| 7                     | 78                          | 5682                            | 151,280                     | 11.4                 |
| 7                     | 85                          | 4,920                           | 156,200                     | 11.45                |
| 7                     | 92                          | 3,150                           | 159,350                     | 11.35                |

Table A-13. Leaching data for Specimen E-2 containing 9.05 wt % iodine as barium iodate plus 0.5% sodium silicate

Curing time = 100 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| <u>Leaching time (days)</u> |                             | <u>Iodine leached(μg)</u> |                             | Final pH of leachate |
|-----------------------------|-----------------------------|---------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ )       | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )     | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                           | 1                           | 6,858                     | 6,858                       | 11.2                 |
| 1                           | 2                           | 7,692                     | 14,550                      | 10.2                 |
| 1                           | 3                           | 7,080                     | 21,630                      | 11.0                 |
| 1                           | 4                           | 6,780                     | 28,410                      | 10.2                 |
| 3                           | 7                           | 9,612                     | 38,022                      | 10.5                 |
| 1                           | 8                           | 3,000                     | 41,022                      | 10.55                |
| 1                           | 9                           | 4,146                     | 45,168                      | 10.6                 |
| 1                           | 10                          | 3,852                     | 49,020                      | 9.9                  |
| 1                           | 11                          | 3,720                     | 52,740                      | 10.6                 |
| 3                           | 14                          | 7,935                     | 60,675                      | 11.1                 |
| 1                           | 15                          | 3,300                     | 63,975                      | 10.8                 |
| 1                           | 16                          | 3,186                     | 67,161                      | 11.25                |
| 1                           | 17                          | 3,150                     | 70,311                      | 11.1                 |
| 1                           | 18                          | 2,763                     | 73,074                      | 10.35                |
| 3                           | 21                          | 6,300                     | 79,374                      | 11.35                |
| 1                           | 22                          | 2,343                     | 81,717                      | 10.4                 |
| 1                           | 23                          | 1,950                     | 83,667                      | 10.2                 |
| 1                           | 24                          | 2,469                     | 86,136                      | 11.0                 |
| 4                           | 28                          | 6,494                     | 92,630                      | 11.6                 |
| 1                           | 29                          | 2,172                     | 94,802                      | 11.1                 |
| 1                           | 30                          | 2,145                     | 96,947                      | 11.1                 |
| 1                           | 31                          | 2,160                     | 99,107                      | 10.2                 |
| 1                           | 32                          | 2,232                     | 101,339                     | 11.1                 |
| 3                           | 35                          | 5,040                     | 106,379                     | 10.9                 |
| 1                           | 36                          | 2,448                     | 108,827                     | 10.9                 |
| 7                           | 43                          | 7,590                     | 116,417                     | 11.5                 |
| 7                           | 50                          | 7,830                     | 124,247                     | 10.6                 |
| 7                           | 57                          | 8,280                     | 132,527                     | 11.0                 |
| 7                           | 64                          | 7,560                     | 140,087                     | 11.1                 |
| 7                           | 71                          | 7,320                     | 147,407                     | 10.9                 |
| 7                           | 78                          | 7,092                     | 154,499                     | 11.4                 |
| 7                           | 85                          | 5,640                     | 160,139                     | 11.45                |
| 7                           | 92                          | 4,230                     | 164,369                     | 11.35                |

Table A-14. Leaching data for Specimen F-1 containing 9.05 wt % iodine as barium iodate plus 0.5% fluosilicic acid

Curing time = 100 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| <u>Leaching time (days)</u>     |                                       | <u>Iodine leached(μg)</u>       |                                       | Final pH of leachate |
|---------------------------------|---------------------------------------|---------------------------------|---------------------------------------|----------------------|
| <u>Incremental</u><br>( $t_n$ ) | <u>Cumulative</u><br>( $\Sigma t_n$ ) | <u>Incremental</u><br>( $a_n$ ) | <u>Cumulative</u><br>( $\Sigma a_n$ ) |                      |
| 1                               | 1                                     | 9,768                           | 9,768                                 | 11.15                |
| 1                               | 2                                     | 4,836                           | 14,604                                | 9.1                  |
| 1                               | 3                                     | 5,172                           | 19,776                                | 11.0                 |
| 1                               | 4                                     | 3,510                           | 23,286                                | 10.2                 |
| 3                               | 7                                     | 7,440                           | 30,726                                | 10.5                 |
| 1                               | 8                                     | 2,208                           | 32,934                                | 10.3                 |
| 1                               | 9                                     | 3,735                           | 36,669                                | 10.65                |
| 1                               | 10                                    | 3,369                           | 40,038                                | 9.8                  |
| 1                               | 11                                    | 3,384                           | 43,422                                | 10.6                 |
| 3                               | 14                                    | 7,200                           | 50,622                                | 10.8                 |
| 1                               | 15                                    | 3,210                           | 53,832                                | 10.6                 |
| 1                               | 16                                    | 2,892                           | 56,724                                | 11.25                |
| 1                               | 17                                    | 2,943                           | 59,667                                | 11.2                 |
| 1                               | 18                                    | 2,616                           | 62,283                                | 10.35                |
| 3                               | 21                                    | 5,880                           | 68,163                                | 11.5                 |
| 1                               | 22                                    | 2,280                           | 70,443                                | 10.3                 |
| 1                               | 23                                    | 2,283                           | 72,726                                | 10.0                 |
| 1                               | 24                                    | 2,469                           | 75,195                                | 11.0                 |
| 4                               | 28                                    | 6,494                           | 91,689                                | 11.6                 |
| 1                               | 29                                    | 1,851                           | 83,540                                | 11.2                 |
| 1                               | 30                                    | 1,974                           | 85,514                                | 11.15                |
| 1                               | 31                                    | 2,070                           | 87,584                                | 10.6                 |
| 1                               | 32                                    | 2,142                           | 89,726                                | 11.1                 |
| 3                               | 35                                    | 4,860                           | 94,586                                | 10.85                |
| 1                               | 36                                    | 2,241                           | 96,827                                | 10.6                 |
| 7                               | 43                                    | 9,420                           | 106,247                               | 11.55                |
| 7                               | 50                                    | 9,180                           | 115,427                               | 10.5                 |
| 7                               | 57                                    | 8,004                           | 123,431                               | 11.2                 |
| 7                               | 64                                    | 7,275                           | 130,706                               | 11.2                 |
| 7                               | 71                                    | 6,858                           | 137,564                               | 10.9                 |
| 7                               | 78                                    | 6,615                           | 144,179                               | 11.2                 |
| 7                               | 85                                    | 6,720                           | 150,899                               | 11.5                 |
| 7                               | 92                                    | 6,690                           | 157,589                               | 11.5                 |

Table A-15. Leaching data for Specimen F-2 containing 9.05 wt % iodine as barium iodate plus 0.5% fluosilicic acid

Curing time = 100 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| <u>Leaching time (days)</u>           |   | <u>Iodine leached (<math>\mu\text{g}</math>)</u> |   | Final pH of leachate |
|---------------------------------------|---|--|---|----------------------|
| <u>Incremental (<math>t_n</math>)</u> | <u>Cumulative (<math>\Sigma t_n</math>)</u> | <u>Incremental (<math>a_n</math>)</u>            | <u>Cumulative (<math>\Sigma a_n</math>)</u> |                      |
| 1                                     | 1   | 9,612  | 9,612                                       | 11.2                 |
| 1                                     | 2   | 6,420  | 16,032                                      | 9.1                  |
| 1                                     | 3   | 2,550  | 18,582                                      | 11.1                 |
| 1                                     | 4   | 1,818  | 20,400                                      | 10.0                 |
| 3                                     | 7   | 6,480  | 26,880                                      | 10.0                 |
| 1                                     | 8   | 630  | 27,510                                      | 10.6                 |
| 1                                     | 9   | 2,346  | 29,856                                      | 10.6                 |
| 1                                     | 10  | 1,995  | 31,851                                      | 9.8                  |
| 1                                     | 11  | 1,995  | 33,846                                      | 10.65                |
| 3                                     | 14  | 4,500  | 38,346                                      | 10.8                 |
| 1                                     | 15  | 1,821  | 40,167                                      | 10.65                |
| 1                                     | 16  | 1,932  | 42,099                                      | 11.3                 |
| 1                                     | 17  | 1,809  | 43,908                                      | 11.1                 |
| 1                                     | 18  | 1,809  | 45,717                                      | 10.25                |
| 3                                     | 21  | 3,615  | 49,332                                      | 11.45                |
| 1                                     | 22  | 1,293  | 50,625                                      | 10.1                 |
| 1                                     | 23  | 1,326  | 51,951                                      | 9.95                 |
| 1                                     | 24  | 1,419  | 53,370                                      | 11.0                 |
| 4                                     | 28  | 3,735  | 57,105                                      | 11.6                 |
| 1                                     | 29  | 1,146  | 58,251                                      | 11.2                 |
| 1                                     | 30  | 1,062  | 59,313                                      | 11.1                 |
| 1                                     | 31  | 1,134  | 60,447                                      | 10.6                 |
| 1                                     | 32  | 1,056  | 61,503                                      | 11.15                |
| 3                                     | 35  | 2,424  | 63,927                                      | 10.85                |
| 1                                     | 36  | 960  | 64,887                                      | 10.65                |
| 7                                     | 43  | 4,704  | 69,591                                      | 11.45                |
| 7                                     | 50  | 4,941  | 74,532                                      | 10.5                 |
| 7                                     | 57  | 3,420  | 77,952                                      | 11.2                 |
| 7                                     | 64  | 2,826  | 80,778                                      | 11.1                 |
| 7                                     | 71  | 2,586  | 83,364                                      | 11.0                 |
| 7                                     | 78  | 2,766  | 86,130                                      | 11.2                 |
| 7                                     | 85  | 2,040  | 88,170                                      | 11.4                 |
| 7                                     | 92  | 1,800  | 89,970                                      | 11.2                 |

Table A-16. Leaching data for Specimen G-1 containing 9.05 wt % iodine as barium iodate plus 0.9% butyl stearate

Cuting time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.6 g

Specimen  
 Mass (M) = 183.4 g<sup>3</sup>  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 7,035                            | 7,035                       | 11.05                |
| 1                     | 2                           | 6,480                            | 13,515                      | 10.1                 |
| 1                     | 3                           | 5,058                            | 18,573                      | 10.8                 |
| 1                     | 4                           | 3,270                            | 21,843                      | 10.1                 |
| 3                     | 7                           | 4,692                            | 26,535                      | 10.3                 |
| 1                     | 8                           | 1,710                            | 28,245                      | 10.4                 |
| 1                     | 9                           | 1,815                            | 30,060                      | 10.65                |
| 1                     | 10                          | 1,686                            | 31,746                      | 9.6                  |
| 1                     | 11                          | 1,704                            | 33,450                      | 9.7                  |
| 3                     | 14                          | 3,321                            | 36,771                      | 10.6                 |
| 1                     | 15                          | 1,212                            | 37,983                      | 10.2                 |
| 1                     | 16                          | 912                              | 38,895                      | 11.25                |
| 1                     | 17                          | 1,473                            | 40,368                      | 10.7                 |
| 1                     | 18                          | 1,200                            | 41,568                      | 10.1                 |
| 3                     | 21                          | 2,484                            | 44,052                      | 11.0                 |
| 1                     | 22                          | 813                              | 44,865                      | 9.6                  |
| 1                     | 23                          | 843                              | 45,708                      | 9.55                 |
| 1                     | 24                          | 1,242                            | 46,950                      | 10.0                 |
| 4                     | 28                          | 2,304                            | 49,254                      | 11.3                 |
| 1                     | 29                          | 552                              | 49,806                      | 10.6                 |
| 1                     | 30                          | 678                              | 50,484                      | 10.6                 |
| 1                     | 31                          | 705                              | 51,189                      | 10.0                 |
| 1                     | 32                          | 726                              | 51,915                      | 10.7                 |
| 3                     | 35                          | 1,575                            | 53,490                      | 10.4                 |
| 1                     | 36                          | 770                              | 54,260                      | 10.25                |
| 7                     | 43                          | 3,540                            | 57,800                      | 11.2                 |
| 7                     | 50                          | 3,495                            | 61,295                      | 10.1                 |
| 7                     | 57                          | 2,766                            | 64,061                      | 10.8                 |
| 7                     | 64                          | 2,400                            | 66,461                      | 10.9                 |
| 7                     | 71                          | 2,352                            | 68,813                      | 10.8                 |
| 7                     | 78                          | 2,280                            | 71,093                      | 11.2                 |
| 7                     | 85                          | 2,295                            | 73,388                      | 11.2                 |
| 7                     | 92                          | 2,220                            | 75,608                      | 10.8                 |

Table A-17. Leaching data for Specimen G-2 containing 9.05 wt % iodine as barium iodate plus 0.9% butyl stearate

Curing time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.6 g

Specimen  
 Mass (M) = 183.4 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma t_n$ ) |                      |
| 1                     | 1                           | 6,846                            | 6,846                       | 11.05                |
| 1                     | 2                           | 6,240                            | 13,086                      | 10.1                 |
| 1                     | 3                           | 5,532                            | 18,618                      | 10.8                 |
| 1                     | 4                           | 3,252                            | 21,870                      | 9.8                  |
| 3                     | 7                           | 4,092                            | 25,962                      | 10.25                |
| 1                     | 8                           | 864                              | 26,826                      | 10.35                |
| 1                     | 9                           | 1,758                            | 28,584                      | 10.6                 |
| 1                     | 10                          | 1,506                            | 30,090                      | 9.6                  |
| 1                     | 11                          | 1,995                            | 32,085                      | 9.8                  |
| 3                     | 14                          | 3,546                            | 35,631                      | 10.6                 |
| 1                     | 15                          | 1,149                            | 36,780                      | 10.25                |
| 1                     | 16                          | 924                              | 37,704                      | 10.25                |
| 1                     | 17                          | 1,383                            | 39,087                      | 10.7                 |
| 1                     | 18                          | 1,089                            | 40,176                      | 10.1                 |
| 3                     | 21                          | 2,184                            | 42,360                      | 11.0                 |
| 1                     | 22                          | 726                              | 43,086                      | 9.6                  |
| 1                     | 23                          | 723                              | 43,809                      | 9.5                  |
| 1                     | 24                          | 900                              | 44,709                      | 10.0                 |
| 4                     | 28                          | 2,100                            | 46,809                      | 11.3                 |
| 1                     | 29                          | 816                              | 47,625                      | 10.6                 |
| 1                     | 30                          | 654                              | 48,279                      | 10.6                 |
| 1                     | 31                          | 771                              | 49,050                      | 10.1                 |
| 1                     | 32                          | 726                              | 49,776                      | 10.7                 |
| 3                     | 35                          | 1,500                            | 51,276                      | 11.4                 |
| 1                     | 36                          | 729                              | 52,005                      | 10.25                |
| 7                     | 43                          | 3,426                            | 55,431                      | 11.2                 |
| 7                     | 50                          | 3,435                            | 58,866                      | 10.1                 |
| 7                     | 57                          | 2,838                            | 61,704                      | 10.6                 |
| 7                     | 64                          | 2,400                            | 64,104                      | 10.9                 |
| 7                     | 71                          | 2,400                            | 66,504                      | 10.8                 |
| 7                     | 78                          | 2,304                            | 68,808                      | 11.2                 |
| 7                     | 85                          | 2,241                            | 71,049                      | 11.2                 |
| 7                     | 92                          | 2,214                            | 73,263                      | 10.8                 |

Table A-18. Leaching data for Specimen H-1 containing 9.05 wt % iodine as barium iodate plus 1.2% polybutene 24 and 0.5% Aerosol OT

Curing time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184.7 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|---------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )           | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,156                           | 6,156                       | 11.15                |
| 1                     | 2                           | 8,892                           | 15,048                      | 10.25                |
| 1                     | 3                           | 7,932                           | 22,980                      | 11.2                 |
| 1                     | 4                           | 4,230                           | 27,210                      | 9.85                 |
| 3                     | 7                           | 4,920                           | 32,130                      | 10.4                 |
| 1                     | 8                           | 3,600                           | 35,730                      | 10.5                 |
| 1                     | 9                           | 4,146                           | 39,876                      | 10.95                |
| 1                     | 10                          | 3,921                           | 43,797                      | 9.9                  |
| 1                     | 11                          | 3,795                           | 47,592                      | 10.4                 |
| 3                     | 14                          | 7,761                           | 55,353                      | 11.6                 |
| 1                     | 15                          | 3,126                           | 58,479                      | 10.7                 |
| 1                     | 16                          | 3,180                           | 61,659                      | 10.25                |
| 1                     | 17                          | 2,892                           | 64,551                      | 11.1                 |
| 1                     | 18                          | 2,616                           | 67,167                      | 11.0                 |
| 3                     | 21                          | 2,826                           | 69,993                      | 11.5                 |
| 1                     | 22                          | 2,226                           | 72,219                      | 10.4                 |
| 1                     | 23                          | 2,133                           | 74,352                      | 10.25                |
| 1                     | 24                          | 2,346                           | 76,698                      | 11.0                 |
| 4                     | 28                          | 5,886                           | 82,584                      | 11.6                 |
| 1                     | 29                          | 2,130                           | 84,714                      | 11.15                |
| 1                     | 30                          | 1,824                           | 86,538                      | 11.15                |
| 1                     | 31                          | 1,920                           | 88,458                      | 10.6                 |
| 1                     | 32                          | 1,878                           | 90,336                      | 11.15                |
| 3                     | 35                          | 4,260                           | 94,596                      | 10.9                 |
| 1                     | 36                          | 2,283                           | 96,879                      | 10.9                 |
| 7                     | 43                          | 16,320                          | 113,199                     | 11.5                 |
| 7                     | 50                          | 11,880                          | 125,079                     | 10.6                 |
| 7                     | 57                          | 6,132                           | 131,211                     | 11.1                 |
| 7                     | 64                          | 5,880                           | 137,091                     | 11.0                 |
| 7                     | 71                          | 5,040                           | 142,131                     | 11.0                 |
| 7                     | 78                          | 4,728                           | 146,859                     | 11.3                 |
| 7                     | 85                          | 4,140                           | 150,999                     | 11.6                 |
| 7                     | 92                          | 4,350                           | 155,349                     | 11.5                 |

Table A-19. Leaching data for Specimen H-2 containing 9.05wt % iodine as barium iodate plus 1.2% polybutene 24 and 0.5% Aerosol OT

Curing time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 16.7 g

Specimen  
 Mass (M) = 184.7 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached( $\mu$ g) |                             | Final pH of leachate) |
|-----------------------|-----------------------------|--------------------------|-----------------------------|-----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )    | Cumulative ( $\Sigma a_n$ ) |                       |
| 1                     | 1                           | 6,135                    | 6,135                       | 11.15                 |
| 1                     | 2                           | 9,360                    | 15,495                      | 10.2                  |
| 1                     | 3                           | 9,000                    | 24,495                      | 11.1                  |
| 1                     | 4                           | 7,500                    | 31,995                      | 10.2                  |
| 3                     | 7                           | 4,920                    | 36,915                      | 10.5                  |
| 1                     | 8                           | 3,600                    | 40,515                      | 10.4                  |
| 1                     | 9                           | 4,500                    | 45,015                      | 10.9                  |
| 1                     | 10                          | 4,080                    | 49,095                      | 9.9                   |
| 1                     | 11                          | 4,086                    | 53,181                      | 10.5                  |
| 3                     | 14                          | 7,935                    | 61,116                      | 11.5                  |
| 1                     | 15                          | 3,180                    | 64,296                      | 10.7                  |
| 1                     | 16                          | 2,880                    | 67,176                      | 11.3                  |
| 1                     | 17                          | 2,973                    | 70,149                      | 11.2                  |
| 1                     | 18                          | 2,571                    | 72,720                      | 11.2                  |
| 3                     | 21                          | 2,715                    | 75,435                      | 11.5                  |
| 1                     | 22                          | 2,130                    | 77,565                      | 10.4                  |
| 1                     | 23                          | 2,100                    | 79,665                      | 10.2                  |
| 1                     | 24                          | 2,283                    | 81,948                      | 11.0                  |
| 4                     | 28                          | 5,607                    | 87,555                      | 11.6                  |
| 1                     | 29                          | 1,962                    | 89,517                      | 11.2                  |
| 1                     | 30                          | 1,689                    | 91,206                      | 11.15                 |
| 1                     | 31                          | 1,776                    | 92,982                      | 10.6                  |
| 1                     | 32                          | 1,728                    | 94,710                      | 11.15                 |
| 3                     | 35                          | 3,795                    | 98,505                      | 10.9                  |
| 1                     | 36                          | 1,857                    | 100,362                     | 10.9                  |
| 7                     | 43                          | 14,640                   | 115,002                     | 11.5                  |
| 7                     | 50                          | 10,932                   | 125,934                     | 10.6                  |
| 7                     | 57                          | 5,208                    | 131,142                     | 11.1                  |
| 7                     | 64                          | 4,470                    | 135,612                     | 11.0                  |
| 7                     | 71                          | 3,990                    | 139,602                     | 11.0                  |
| 7                     | 78                          | 3,600                    | 143,202                     | 11.45                 |
| 7                     | 85                          | 3,300                    | 146,502                     | 11.6                  |
| 7                     | 92                          | 3,306                    | 149,808                     | 11.5                  |

Table A-20. Leaching data for Specimen I-1 containing 9.05 wt % iodine as barium iodate plus 1.4% lead monoxide

Curing time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 17.0 g

Specimen  
 Mass (M) = 187.4 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|---------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )           | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,360                           | 6,360                       | 11.3                 |
| 1                     | 2                           | 8,892                           | 15,252                      | 10.2                 |
| 1                     | 3                           | 9,132                           | 24,384                      | 11.3                 |
| 1                     | 4                           | 7,848                           | 32,232                      | 10.1                 |
| 3                     | 7                           | 12,600                          | 44,832                      | 10.55                |
| 1                     | 8                           | 3,252                           | 48,084                      | 10.8                 |
| 1                     | 9                           | 5,826                           | 53,910                      | 10.9                 |
| 1                     | 10                          | 5,460                           | 59,370                      | 10.1                 |
| 1                     | 11                          | 5,355                           | 64,725                      | 10.7                 |
| 3                     | 14                          | 9,390                           | 74,115                      | 12.0                 |
| 1                     | 15                          | 4,446                           | 78,561                      | 10.7                 |
| 1                     | 16                          | 4,200                           | 82,761                      | 11.3                 |
| 1                     | 17                          | 3,789                           | 86,550                      | 11.2                 |
| 1                     | 18                          | 3,966                           | 90,516                      | 11.3                 |
| 3                     | 21                          | 8,001                           | 98,517                      | 11.6                 |
| 1                     | 22                          | 3,192                           | 101,709                     | 10.3                 |
| 1                     | 23                          | 3,150                           | 104,859                     | 10.3                 |
| 1                     | 24                          | 3,270                           | 108,129                     | 11.25                |
| 4                     | 28                          | 8,466                           | 116,595                     | 11.8                 |
| 1                     | 29                          | 3,000                           | 119,595                     | 11.3                 |
| 1                     | 30                          | 2,718                           | 122,313                     | 11.3                 |
| 1                     | 31                          | 2,793                           | 125,106                     | 10.7                 |
| 1                     | 32                          | 2,910                           | 128,016                     | 11.35                |
| 3                     | 35                          | 6,360                           | 134,376                     | 11.0                 |
| 1                     | 36                          | 3,000                           | 137,376                     | 11.0                 |
| 7                     | 43                          | 12,000                          | 149,376                     | 11.8                 |
| 7                     | 50                          | 4,026                           | 153,402                     | 10.9                 |
| 7                     | 57                          | 9,360                           | 162,762                     | 11.3                 |
| 7                     | 64                          | 8,520                           | 171,282                     | 11.1                 |
| 7                     | 71                          | 7,470                           | 178,752                     | 11.2                 |
| 7                     | 78                          | 7,320                           | 186,072                     | 11.5                 |
| 7                     | 85                          | 10,368                          | 196,440                     | 11.7                 |
| 7                     | 92                          | 4,800                           | 201,240                     | 11.5                 |

Table A-21. Leaching data for Specimen I-2 containing 9.05wt % iodine as barium iodate plus 1.4% lead monoxide

Curing time = 34 days  
 Leachant = 300 ml distilled water, pH = 7  
 Iodine content ( $A_0$ ) = 17.0 g

Specimen  
 Mass (M) = 187.4 g  
 Volume (V) = 98.2 cm<sup>3</sup>  
 Exposed area (S) = 19.64 cm<sup>2</sup>

| Leaching time (days)  |                             | Iodine leached ( $\mu\text{g}$ ) |                             | Final pH of leachate |
|-----------------------|-----------------------------|----------------------------------|-----------------------------|----------------------|
| Incremental ( $t_n$ ) | Cumulative ( $\Sigma t_n$ ) | Incremental ( $a_n$ )            | Cumulative ( $\Sigma a_n$ ) |                      |
| 1                     | 1                           | 6,495                            | 6,495                       | 11.3                 |
| 1                     | 2                           | 10,236                           | 16,731                      | 10.25                |
| 1                     | 3                           | 9,888                            | 26,619                      | 11.2                 |
| 1                     | 4                           | 7,962                            | 34,581                      | 10.25                |
| 3                     | 7                           | 13,128                           | 47,709                      | 10.5                 |
| 1                     | 8                           | 4,488                            | 52,197                      | 10.55                |
| 1                     | 9                           | 6,000                            | 58,197                      | 10.95                |
| 1                     | 10                          | 5,295                            | 63,492                      | 10.1                 |
| 1                     | 11                          | 5,541                            | 69,033                      | 10.7                 |
| 3                     | 14                          | 9,132                            | 78,165                      | 11.8                 |
| 1                     | 15                          | 5,364                            | 83,529                      | 10.75                |
| 1                     | 16                          | 4,266                            | 87,795                      | 11.25                |
| 1                     | 17                          | 4,203                            | 91,998                      | 11.2                 |
| 1                     | 18                          | 4,350                            | 96,348                      | 11.35                |
| 3                     | 21                          | 7,866                            | 104,214                     | 11.6                 |
| 1                     | 22                          | 3,246                            | 107,460                     | 10.4                 |
| 1                     | 23                          | 3,156                            | 110,616                     | 10.3                 |
| 1                     | 24                          | 3,330                            | 113,946                     | 11.25                |
| 4                     | 28                          | 8,286                            | 122,232                     | 11.8                 |
| 1                     | 29                          | 3,180                            | 125,412                     | 11.3                 |
| 1                     | 30                          | 2,718                            | 128,130                     | 11.3                 |
| 1                     | 31                          | 2,793                            | 130,923                     | 10.75                |
| 1                     | 32                          | 2,910                            | 133,833                     | 11.4                 |
| 3                     | 35                          | 6,300                            | 140,133                     | 11.0                 |
| 1                     | 36                          | 3,216                            | 143,349                     | 11.0                 |
| 7                     | 43                          | 9,648                            | 152,997                     | 11.8                 |
| 7                     | 50                          | 3,600                            | 156,597                     | 10.9                 |
| 7                     | 57                          | 9,528                            | 166,125                     | 11.3                 |
| 7                     | 64                          | 8,448                            | 174,573                     | 11.1                 |
| 7                     | 71                          | 8,052                            | 182,625                     | 11.2                 |
| 7                     | 78                          | 8,424                            | 191,049                     | 11.55                |
| 7                     | 85                          | 6,402                            | 197,451                     | 11.7                 |
| 7                     | 92                          | 4,110                            | 201,561                     | 11.5                 |

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