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**EFFECT OF EXTENDED EXPOSURE TO
SIMULATED LMFBR FUEL
REPROCESSING OFF-GAS ON
RADIOIODINE TRAPPING PERFORMANCE OF
SORBENTS (FINAL REPORT)**

R. D. Ackley

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CHEMICAL TECHNOLOGY DIVISION

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ON RADIOIODINE TRAPPING PERFORMANCE OF SORBENTS (FINAL REPORT)

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AUGUST 1974

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CONTENTS

	Page
ABSTRACT	1
1. SUMMARY	1
2. INTRODUCTION	4
3. EXPERIMENTAL	6
3.1 Apparatus and Operating Conditions	6
3.2 Oxidizing Catalyst and Sorbents	11
3.3 Procedure	12
3.4 Radioactivity Measurements	12
3.5 Evaluation of Decontamination Factors	13
4. RESULTS AND DISCUSSION.	15
4.1 Decay Effects	15
4.2 Exchange Effects	16
4.3 Observed Decontamination Factors for Iodine and Methyl Iodide	18
4.4 Trapping Behavior of Elemental Iodine and Methyl Iodide	28
4.5 Concerning the Desirability of Using a Catalytic Oxidation Bed	29
4.6 Sorbent Shape and Particle Size	30
5. ACKNOWLEDGMENTS	31
6. REFERENCES	32

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R. D. Ackley and R. J. Davis

ABSTRACT

The effect of extended exposure to simulated LMFBR fuel reprocessing off-gas on the radioiodine trapping performance of certain sorbents was investigated. Satisfactory trapping performance for iodine injected as CH_3I or as I_2 was observed for the following sorbent--operating temperature combinations over exposure times of ≥ 195 days and for the case of the simulated off-gas not having been specially treated: 1/16-in. pellets of 26% AgX (silver-sodium zeolite with 26% Ag^+ for Na^+ exchange) at 200°C ; 1/16-in. pellets of 99% AgX at 100°C ; and 8 x 16 mesh (U.S.) granules of GX135 (silver salt-treated alumina-silica) at 100°C . In less-extended testing, satisfactory performance was also observed for 10 x 20 mesh beads of 88% AgX at 200°C . Results were also obtained for the case of exposure to simulated off-gas which had been treated by passage through Hopcalite, an oxidizing catalyst, at 350°C ; both beneficial and detrimental effects were observed.

1. SUMMARY

A required radioiodine retention factor of $\sim 10^8$ is projected for LMFBR fuel reprocessing plants handling short-decayed fuel. Also projected is a cumulative retention factor of $\sim 10^6$ for iodine fixation achieved upstream of the filter-adsorber assembly used for final off-gas treatment. Thus, this assembly will need to provide a decontamination factor (DF) of $\sim 10^2$, but considerable reserve capability is desired. Certain silver-containing sorbents in a 2-in. depth at 100°C and/or 200°C have, in short-term laboratory tests,¹ exhibited DFs of $> 10^3$ for iodine as CH_3I or as I_2 under conditions including 3 vol % water vapor and iodine loadings of not less than 5 mg per cm^3 of sorbent. The more recent objectives were to obtain information on the effective service life of such sorbents and to ascertain the desirability of locating a catalytic oxidation bed upstream of the sorbents to minimize poisoning by organics and to convert organic iodides to I_2 .

Accordingly, a laboratory facility for exposing sorbents to simulated fuel reprocessing off-gas was constructed and operated. The

main air stream was humidified to $\sim 85\%$ saturation (referred to room temperature). Elemental iodine labeled with ^{125}I ($t_{1/2} = 60$ days), dodecane, and nitric oxide were injected, generally on a continuous basis, at or reasonably near their respective target concentrations. One group of sorbents was exposed to untreated simulated off-gas while another group was exposed to simulated off-gas which had traversed a Hopcalite bed at 350°C . Methyl iodide labeled with ^{130}I ($t_{1/2} = 12.3$ hr) was injected periodically to provide the means for obtaining short-term trapping results, whereas long-term results were obtained with the I_2 .

Certain of the DF results obtained may have reflected the effects of exchange processes which, in general, would tend to enhance the DF values. Corrections for such effects were not actually applied due to the uncertainty with regard to the degree to which these effects were operative. In treating the data, the effects of radioactive decay on the DF values were eliminated by appropriate correction. In an actual off-gas application, decay effects would aid in achieving good iodine retention performance.

The sorbents investigated were four varieties of silver zeolite or, alternatively, silver-sodium zeolite (to be designated AgX with the percent Ag^+ for Na^+ exchange denoted), GX135 (silver salt-treated alumina-silica), and a type of iodized charcoal. A sorbent depth of 2 in. was used in each case.

Satisfactory performance (CH_3I DFs $\geq 9 \times 10^3$ and I_2 DFs $\geq 5 \times 10^2$) was observed for the following test situations over the exposure times indicated which, for each test situation, are with reference to CH_3I and I_2 , respectively:

- (1a) 26% AgX, 1/16-in. pellets, at 200°C , without Hopcalite upstream, exposure times of 286 and 350 days. (Final CH_3I value obtained was at 286 days; for (1a) through (5b), the second number of days also represents the overall exposure period for that test situation.)
- (1b) As in (1a) except that Hopcalite was upstream.
- (2a) GX135, 8 x 16 mesh (U.S.) granules, at 200°C , without Hopcalite upstream, exposure times of 77 and 350 days. (Performance for CH_3I deteriorated significantly between 77 and 140 days.)

- (2b) GX135, at 200°C, with Hopcalite upstream, exposure times of 286 and 350 days. (Final CH₃I value was at 286 days.)
- (3a) GX135, at 100°C, without Hopcalite upstream, exposure times of 209 and 273 days. (Final CH₃I value was at 209 days.)
- (3b) GX135, at 100°C, with Hopcalite upstream, exposure times of 90 and 154 days. (Only CH₃I value obtained was at 90 days; overall exposure time was relatively short.)
- (4a) 99% AgX, 1/16-in. pellets, at 100°C, without Hopcalite upstream, exposure times of 195 and 259 days. (Final CH₃I value was at 195 days.)
- (4b) As in (4a) except that Hopcalite was upstream.
- (5a) 88% AgX, 10 x 20 mesh (U.S.) beads, at 200°C, without Hopcalite upstream, exposure times of 90 and 154 days. (Only CH₃I value obtained was at 90 days; overall exposure time was relatively short.)
- (5b) As in (5a) except that Hopcalite was upstream.

The performance observed for the type of iodized charcoal that was included mainly for purposes of comparison with previous experience indicated that its useful service life under conditions similar to those employed would be limited to 60 days or less. A striking feature of the results for this sorbent was that trapping of iodine introduced as I₂ was adversely affected to a pronounced degree by the use of Hopcalite. This behavior, together with related observations, is discussed in some detail in Sect. 4.4.

The performance observed for 88% silver zeolite in the form of 1/8-in. pellets at both 100°C and 200°C was only fair relative to that for 1/16-in. pellets of 26% AgX or 10 x 20 mesh beads of 88% AgX at 200°C, or 1/16-in. pellets of 99% AgX at 100°C. This illustrates the importance of sorbent particle size.

In general, the CH₃I and I₂ DFs observed for the sorbents exhibiting satisfactory performance were relatively high and at or near the limit of the analytical resolution regardless of whether the gas stream was pretreated by a Hopcalite bed. Thus, these particular results, taken as a whole, were inconclusive regarding the value of pretreating the

off-gas. However, they do tend to indicate that the possible benefit to be gained by the use of a catalytic oxidation bed in the off-gas system would probably not be of sufficient magnitude to warrant the associated added cost.

Other investigators have pointed out that granular sorbents are more effective than those in the form of pellets or beads.² Obviously, this aspect, as well as the one bearing on optimum particle size, should be given careful consideration when a sorbent is selected for actual application.

2. INTRODUCTION

In developing the technology for LMFBR fuel reprocessing plants, radioiodine control represents a major challenge since large amounts of short-cooled fuel will be involved. A required radioiodine retention factor of $\sim 10^8$ (overall) is projected for an appropriately-sized plant. Also projected is a cumulative retention factor of $\sim 10^6$ corresponding to iodine removal achieved upstream of the final off-gas trapping system. The latter value is based on consideration of iodine fixation resulting from mechanisms inherent in the fuel reprocessing operations and on evaluation of the expected performance of iodine removal equipment (exclusive of the final trapping system) incorporated in the reprocessing facility. Accordingly, the filter-adsorber assembly employed for final treatment of the off-gas will need to provide a radioiodine decontamination factor (DF) of $\sim 10^2$; however, considerable reserve capability would, of course, be desirable. The overall problem of iodine retention in LMFBR fuel reprocessing plants, as outlined above, has been discussed in detail by Yarbrow, Crouse, and Cathers.³ A rather extensive investigative program on iodine retention is being conducted in the Chemical Technology Division of ORNL as a part of a larger program on LMFBR fuel reprocessing.⁴ The work described herein was performed in conjunction with this iodine retention program and, more specifically, was concerned with determining the applicability of various sorbents for trapping iodine in the off-gas system employed for final treatment.

Certain silver-containing sorbents in a 2-in. depth at 100°C and/or 200°C have, in short-term laboratory tests,¹ exhibited DFs of $> 10^3$ for iodine either as CH_3I or as I_2 under conditions which included 3 vol % water vapor in the carrier gas (air), a face velocity of 40 fpm referred to 25°C, and iodine loadings of not less than 5 mg per cm^3 of sorbent. Two of these effective sorbents were prepared in the laboratory and are referred to as silver zeolites (or, alternatively, as silver-sodium zeolites) with the extent of Ag^+ for Na^+ exchange being approximately 27% and 90%. The selection of these sorbents for investigation in connection with treating LMFBR fuel reprocessing off-gas was prompted by the development reported by Maeck, Pence, and Keller,⁵ namely that silver zeolite is highly effective for trapping iodine in the forms of CH_3I and I_2 . (They mentioned the possible potential of silver zeolite for removing iodine from fuel reprocessing off-gas.) Another effective material was a silver-containing inorganic sorbent, GX135, obtained from a commercial source.

While silver-containing sorbents are expensive, the quantity of off-gas to be treated is projected to be relatively small (~ 500 cfm for a typical plant); therefore, extremely large quantities of an effective sorbent will probably not be required. Furthermore, if the off-gas flow rate and the sorbent volume are both small, the use of heated sorbent beds to decrease relative humidity and thus improve trapping efficiency is well within the realm of feasibility.

The main objective was to obtain information on the effective service life of these promising sorbents. The secondary objective was to ascertain the desirability of locating a catalytic oxidation bed upstream of the filter-adsorber assembly. The oxidation bed would have two functions: (1) to minimize poisoning of the sorbent by catalytic combustion of adsorbable organic vapors, and (2) to convert iodine in the form of organic iodides to elemental iodine with the expectation of improving trapping efficiency. The only catalyst employed in this work was Hopcalite, and its use in connection with iodine trapping has been investigated previously.^{3,6-9}

This report is intended to supersede an earlier interim report covering a portion of the work described here.¹⁰

3. EXPERIMENTAL

In this investigation, selected sorbents were exposed to simulated LMFBR fuel reprocessing off-gas for extended periods of time which varied, for different sorbent-condition combinations, from 77 to 350 days. One group of sorbents was exposed to untreated off-gas, while a duplicate or near-duplicate group was exposed to off-gas which had traversed a bed of heated Hopcalite. Periodically, trapping data were obtained for iodine injected as I_2 and/or as CH_3I . The experimental modes were such that the results for elemental iodine are basically long term, while those for CH_3I are short term.

3.1 Apparatus and Operating Conditions

Figure 1 shows a schematic diagram of the laboratory facility that was employed. In this figure, A, B, C, D, and E represent what are termed test bed sections, whereas the F's represent the collection bed sections. (More details regarding these sections are given in Table 1.) Certain of the initially installed test sorbents were later replaced, and, in some of these cases, the sorbent temperatures were altered. Also, on occasion and for relatively brief periods, iodine characterization packs of the type developed by Bennett, Hinds, and Adams¹¹ were installed in the facility to sample and analyze the iodine species in the simulated off-gas. Excluding downtime (e.g., for changeout of sorbents or Hopcalite), the facility was operated for 350 days. During the last 154 days of this period, two secondary (so-called) collection bed sections were included, one in parallel with each group of sorbents, to provide data for estimating the amounts of iodine injected as I_2 and penetrating to the test beds. The collection beds indicated in Fig. 1 (items F) will, in this context, be referred to as the primary ones.

In construction of the facility, stainless steel type 316 was employed, almost exclusively, where corrosion might be a problem. The various flowmeters that were utilized, including the ten exit flowmeters for the primary collection beds (and the two for the secondary beds), are not shown. Table 1 gives the dimensions of the sorbent beds and the catalytic oxidation bed, together with certain of the operating conditions such as velocities for the simulated off-gas and approximate or estimated

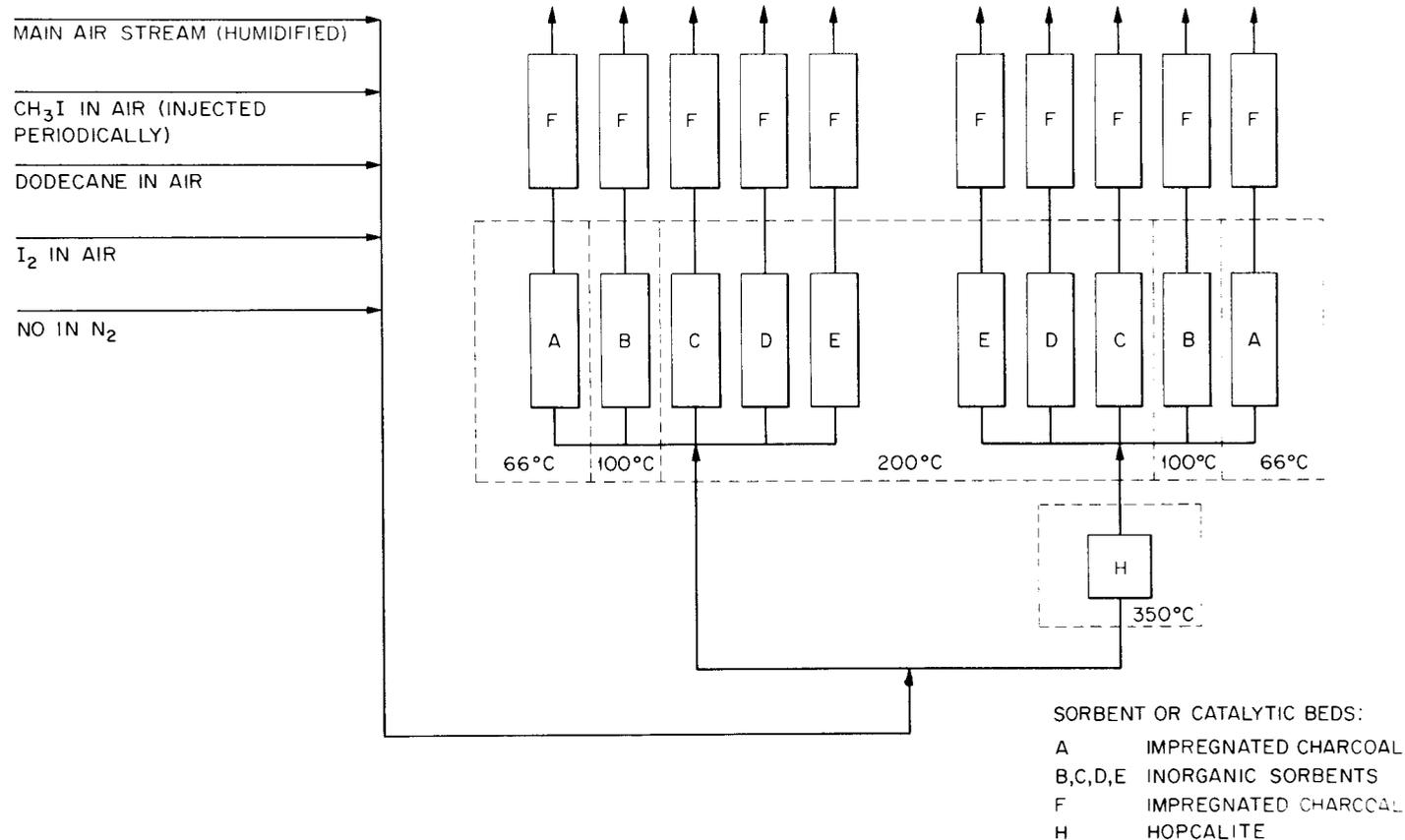


Fig. 1. Flow Diagram of Apparatus for Investigating Effects of Long-Term Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Trapping Performance of Sorbents.

Table 1. Experimental Details for Long-Term Facility

Each test bed section: two 1-in.-deep beds in series, bed diameter of 1.05 in.

Test bed temperatures (mean): 66, 100, or 200°C

Superficial off-gas velocities in test beds: 46, 50, or 63 fpm (respectively, for preceding temperatures)

Each primary collection bed section: three 1-in.-deep beds in series, bed diameter of 1 in.

Each secondary collection bed section: two 1-in.-deep beds in series, bed diameter of 1 in.

Hopcalite bed: 2.6 in. deep, 2.1-in. diameter, 350°C, superficial off-gas velocity of 108 fpm

Main air stream: 68 liters/min at 26°C, 1 atm, ~ 85% R. H. at 26°C

Dodecane concentration (approximate overall average): 1.5×10^{-6} g/liter^a

Nitric oxide concentration (approximate overall average): 1.2×10^{-5} g/liter^a

Estimated average I₂ concentrations at entrances to test beds not downstream of Hopcalite:

First 77 days exposure	9×10^{-8} g/liter ^a
Next 119 days exposure	4×10^{-8} g/liter
Next 29 days exposure	$< 10^{-10}$ g/liter
Next 61 days exposure	14×10^{-8} g/liter
Next 29 days exposure	5×10^{-9} g/liter
Next 35 days exposure	18×10^{-8} g/liter
Time-weighted average for 350 days	8×10^{-8} g/liter

Estimated average I₂ concentrations at entrances to test beds downstream of Hopcalite:

For each of the exposure periods indicated above, ~ 0.8 times the concentration value quoted.

Periodic CH₃I injection: equivalent to ~ 0.1 mg of I per cm³ of sorbent per injection; ~ 2×10^{-4} g of CH₃I/liter^a (over ~ 3-min period).

Radioactive labeling:

I ₂ : with ¹²⁵ I (t _{1/2} = 60 days)	CH ₃ I: with ¹³⁰ I (t _{1/2} = 12.3 hr)
---	---

^aConcentration values quoted above are with respect to the simulated off-gas at approximately 1 atm and 26°C.

concentrations for the additives. The reported temperatures are based on readings exhibited by thermocouples with junctions located, in each case, on the center line immediately downstream of the sorbent or catalyst. Considering the gradients that were present, each such value is regarded as being representative of the mean temperature for the test bed section or Hopcalite bed involved.

During operation of the facility, the main air stream was humidified to $\sim 85\%$ saturation referred to room temperature (near 26°C). Practical-grade dodecane (mp, -10.5 to -9°C)* and technical-grade nitric oxide** were introduced continuously into the main air stream to provide concentrations which, on the basis of long-term usage, were satisfactorily near the respective target concentrations of 2×10^{-6} g/liter and 1×10^{-5} g/liter.

Elemental iodine labeled with ^{125}I ($t_{1/2} = 60$ days) was also continuously injected except for several brief periods of a few days duration or less (e.g., when the supply was exhausted) and except for a 29-day period during which the indicated I_2 concentration was practically zero. Also, during one other 29-day period, it was considerably lower than that desired. To compensate for the low concentration levels that prevailed during the two 29-day periods, relatively high concentration levels were employed during the succeeding periods. The target I_2 concentration range originally specified was 1×10^{-8} to 10×10^{-8} g/liter. (For the conditions involved in this particular investigation, an iodine concentration of, e.g., 4×10^{-8} g/liter is equivalent to a loading rate of 5 mg of iodine per cm^3 of sorbent in a 2-in. depth per year.)

Originally, I_2 was injected into the main air stream via a stainless steel valve and a short section of stainless steel tubing; later, after the first 196 days of operation, injection was made via a Teflon valve and Teflon-lined tubing to minimize I_2 deposition in the side

* Obtained from Matheson Coleman and Bell.

** Obtained from The Matheson Company.

stream. The second injection arrangement proved highly satisfactory, although some time elapsed before consistent operation was achieved.

While different batches of I_2 were injected during the first 196 days of operation, all of them were prepared so as to have nearly the same specific activity at any particular time. This constancy of specific activity had the advantage of allowing the calculation of cumulative decontamination factors for I_2 ; however, after several months, the ^{125}I activity level had become too low to maintain good counting statistics in cases where the DF values were high. Subsequently (i.e., for the last 154 days of operation), three batches of I_2 , each having a rather high initial specific activity, were employed. This modification of technique resulted in the later I_2 DFs being incremental rather than cumulative. Two I_2 batches were injected during the first or 90-day incremental period (of the 154 days), and one batch was injected during the final or 64-day period. (However, the I_2 injection rate was negligible or low during earlier portions of both incremental periods.)

As indicated in Table 1, the I_2 concentrations considered to be of principal interest were those pertaining to the simulated off-gas as it entered the test beds. The concentration values quoted for the first 196 days were estimated by using the amounts of ^{125}I found on removed test beds and their associated primary collection beds; the values for the last 154 days were estimated using the amounts of ^{125}I found on the secondary collection beds in conjunction with the amounts of I_2 introduced into the main air stream. (The assumption that the I_2 concentration under consideration did not exceed 0.9 times that calculated from the amount of I_2 injected was involved in making the latter concentration estimates; also involved was the observation that the ratio of the total of the amounts of ^{125}I found on the secondary collection beds downstream of Hopcalite to the total for those beds not downstream of Hopcalite was approximately 0.8.) The estimated uncertainty in these I_2 concentration values (Table 1) is $\leq 50\%$, with the uncertainty being mainly a consequence of complications encountered in performing ^{125}I assay measurements (as described in Sect. 3.4).

While elemental iodine was injected on a generally continuous basis, ^{130}I -labeled CH_3I was injected only periodically (roughly bimonthly on average) and then practically as a pulse. Injection was probably virtually complete in about 3 min.

3.2 Oxidizing Catalyst and Sorbents

Identification of the catalyst and the sorbents, mean operating temperatures of the test bed sections, status of the latter regarding whether Hopcalite was (HUS) or was not (HNUS) upstream, and information regarding exposure time in the facility are given below.

Oxidizing Catalyst:

Hopcalite, MSA Part No. 21215, 8 x 16 mesh (U.S.), from Mine Safety Appliances Company. Replaced after, referring to successive cases, 77, 63, 56, and 90 days of operation. Final bed was used 64 days.

Sorbents Tested:

Activated carbon, iodized, Grade 42, 8 x 16 mesh (U.S.), from Witco Chemical Corporation. Test bed sections: (1) 66°C, HNUS, first 77 days (of overall exposure time of 350 days); (2) 66°C, HUS, first 196 days.

88% silver zeolite, 1/8-in. pellets, from Materials Systems Division of Union Carbide Corporation. Test bed sections: (1) 100°C, HNUS, first 77 days; (2) as (1) except HUS; (3) 200°C, HNUS, first 196 days; (4) as (3) except HUS.

26% silver zeolite, 1/16-in. pellets, prepared in the laboratory by treating Linde Molecular Sieve Type 13X with AgNO₃ solution. (Target degree of exchange was 25%.) Test bed sections: (1) 200°C, HNUS, 350 days; (2) as (1) except HUS.

GX135, 8 x 16 mesh (U.S.) granules, silver salt-treated alumina-silica, from North American Carbon, Inc. Test bed sections: (1) 200°C, HNUS, 350 days; (2) as (1) except HUS; (3) 100°C, HNUS, last 273 days; (4) 100°C, HUS, last 154 days.

99% silver zeolite, 1/16-in. pellets, from Materials Systems Division of Union Carbide Corporation. Test bed sections: (1) 100°C, HNUS, last 259 days; (2) as (1) except HUS.

88% silver zeolite, 10 x 20 mesh (U.S.) beads, from Davison Chemical Division of W. R. Grace and Company. Test bed sections: (1) 200°C, HNUS, last 154 days; (2) as (1) except HUS.

Collection Bed Sorbents:

Activated carbon, iodized, MSA 85851, 8-14 mesh (Tyler), from Mine Safety Appliances Company. Used in primary collection bed sections for first 286 days with one exception, that being for GX135 at 200°C, HNUS, and during last 90 days of first 286 days; in the latter case, the material used was that described next.

93% silver zeolite, 1/16-in. pellets, prepared in the laboratory (13X-based). Used in primary collection bed sections during last 64 days and in secondary collection bed sections. The latter were maintained at $\sim 100^\circ\text{C}$. The primary collection beds were not externally heated but, as a consequence of having been exposed to gas which had traversed the test beds, were at temperatures moderately above ambient.

3.3 Procedure

Initially, the catalyst (Hopcalite) and sorbents were preequilibrated at temperature with the flowing humidified air for 27 hr. (Each time fresh Hopcalite was employed, it was degassed at 350°C prior to connecting its holder with the line leading to the downstream sorbents.) Next, the first CH_3I injection was performed. Approximately 16 hr later, the collection beds were removed (and replaced) and assayed for ^{130}I . Then, a few hours later, the introduction of dodecane, I_2 , and NO was initiated. In obtaining subsequent CH_3I trapping results, the earlier procedure of injection, allowing 16 to 18 hr to elapse (with facility operating), and removal of the collection beds followed by their ^{130}I assay was repeated. In addition, the collection beds were assayed for ^{125}I after allowing sufficient time for virtually complete decay of ^{130}I . (The introduction of additives continued undisturbed during this subsequent testing with CH_3I .) While CH_3I results were obtained for the test sorbents initially installed prior to the introduction of dodecane, NO , and I_2 , analogous results (i.e., for zero exposure time) were not obtained for those sorbent beds which were subsequently installed. Also, CH_3I data corresponding to the end of the 350-day exposure period were not obtained; however, the results for the ^{125}I -labeled I_2 were obtained as usual.

3.4 Radioactivity Measurements

The relative net counting rates* for the various sorbent beds that were removed, and for the associated screens and their retainers, were measured using a gamma spectrometer equipped with a 3 in. x 3 in. NaI detector in conjunction with a multichannel analyzer. For ^{125}I , the

*The net counting rate is the gross counting rate minus the background counting rate.

analyzer was set to count over the channels including the photopeak at 28 keV and the summation peak at 54 keV.¹² For ¹³⁰I, the photopeaks included were those at 0.42, 0.54, 0.67, and 0.74 MeV.¹³ Decay corrections were applied where necessary.

The selection of ¹²⁵I as the I₂ tracer was based on its relatively long half-life (60 days), which offered an obvious advantage in this type of investigation. However, this advantage was offset, to some extent, by the different degrees of attenuation exhibited by sorbents with differing composition and density, a consequence of the low energy of ¹²⁵I radiation.¹² Allowance was made for this effect by using counting data observed for standard counting samples, which consisted of appropriate volumes of various sorbents impregnated with definite amounts of ¹²⁵I solution. Even so, the I₂ results, which are calculated from counting rates involving different sorbents and differing degrees of intraparticle penetration of ¹²⁵I, probably reflect a certain amount of error due to uncertainties in the corrections for attenuation. However, the average error from this source in the calculated I₂ DF values probably does not exceed $\pm 50\%$, which would appear to represent an adequate degree of accuracy for present purposes.

3.5 Evaluation of Decontamination Factors

The CH₃I results obtained in this investigation are given on a noncumulative basis. Each of the CH₃I DFs associated with a given 2-in. depth of sorbent is, or is based on (referring to values preceded by a "greater than" or "less than" sign), the following quotient: one-tenth of the decay-corrected net counting rate* of the ¹³⁰I-labeled source divided by the sum of the decay-corrected net counting rates found for the parts (beds, etc.) of the collection bed section downstream of that 2-in. depth of sorbent. (The apportionment is one-tenth because there were ten test bed sections.) The CH₃I DFs associated with test sorbents downstream of the Hopcalite are regarded as indicative of I₂ trapping capability since the CH₃I that traversed the Hopcalite bed was presumably converted to elemental iodine.⁷ Some fraction of the latter was undoubtedly retained on surfaces upstream of the test sorbents. Also, considering the manner in which the CH₃I DFs were evaluated, any amounts

*This counting rate is determined just prior to injection.

of ^{130}I retained in the connecting lines between the test bed sections and the collection bed sections were, in effect, included with the quantities trapped further upstream. (The connecting lines were of 1/4-in.-OD, 35-mil-wall, stainless steel type 316; lengths were 0.7 ft in cases where the test bed temperatures were 66 or 100°C and 5 ft in cases where such temperatures were 200°C.) Accordingly, each of these CH_3I DFs corresponds to the overall result of all removal processes which occurred between the point of injection and the point of entrance into the applicable collection bed section.

As contrasted with the results for CH_3I , the I_2 DFs covering the first 196 days of operation were evaluated on a cumulative basis and apply, within the limitations to be indicated, to those quantities of ^{125}I which entered the test bed sections. For each of the cases in which the test beds were removed and assayed for ^{125}I , excluding two cases where the ^{125}I distribution in the associated collection beds was indicative of incomplete trapping, the quantity of iodine entering was assumed to be that found on the removed test beds (the two 1-in. depths in series) plus that found on the two or more successive collection bed sections associated with them. (Actually, the calculations were made in terms of counting rates corrected as already described.) For other cases, the quantities of entering iodine employed were estimates based on those quantities evaluated as described above, length of exposure time, and the particular time period (to allow for the variation in effective I_2 concentration). Thus, each of these I_2 DFs is, or is based on, the quotient obtained by dividing the calculated amount of iodine (injected as I_2) entering the test bed section (during the specific exposure time) by the total amount found on the collection bed section(s) located downstream during that exposure time. (Values preceded by a "greater than" sign include an allowance which tended to reduce the numerical value for the uncertainty due to attenuation effects; the other I_2 DF values do not.) Quantities of iodine retained in the lines between the test bed sections and the collection bed sections were not measured and thus were excluded in evaluating the quantities entering the test bed sections. However, the former were probably negligible relative to the latter (as evaluated

or estimated). Accordingly, each of the I_2 DFs is interpreted as being indicative of the effect of removal processes occurring between the point of entrance to the applicable test bed section and the point of entrance to its associated collection bed section or successive collection bed sections. (That is, the I_2 DFs relate in each case to a more limited portion of the apparatus than do the CH_3I DFs.)

The I_2 DFs for the last 154 days of operation were, in contrast to the preceding for I_2 DFs, evaluated on an incremental basis; the incremental periods involved were 90 and 64 days. (However, the I_2 injection rate was low or negligible during the first 29 days of each of these periods.) The estimated amounts entering the test bed sections during each period were calculated on the basis of the amounts of ^{125}I found on the particular secondary collection beds that were employed during each period. (The mass flow rate for each of the latter beds was 1/6.8 times that for each test bed section.) Otherwise, considerations similar to those corresponding to the earlier I_2 DF values apply. Also, in connection with continued test situations, the I_2 DFs were undoubtedly affected by the presence of previously trapped I_2 having a different specific activity. Some cognizance of this aspect is included in the discussion of the results.

The effects of iodine retention on nonsorbent surfaces may not be as important as is perhaps suggested by certain parts of the above discussion. However, definitive information on this aspect is lacking, and, therefore, recognition of the possible role of nonsorbent surfaces in achieving the DFs obtained in this investigation appears to be necessary.

4. RESULTS AND DISCUSSION

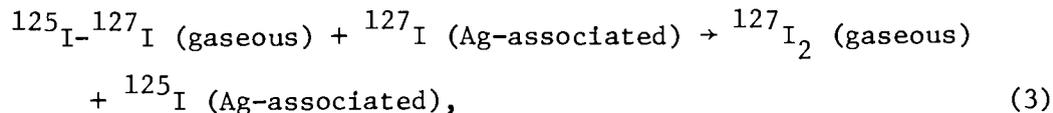
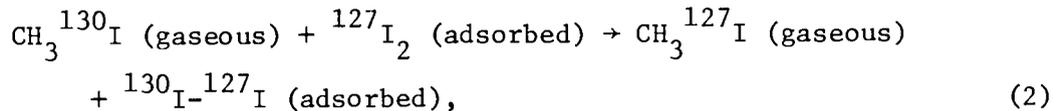
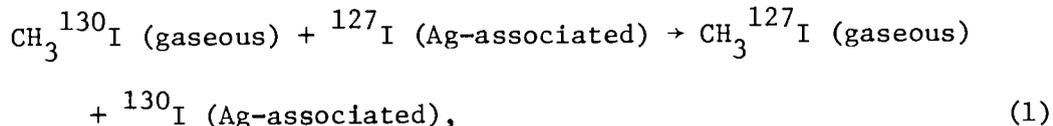
4.1 Decay Effects

In the reprocessing of LMFBR fuel which has cooled for the projected period of 30 days, the iodine isotopes of primary concern will be 1.7×10^7 -year ^{129}I , 8.07-day ^{131}I , and 2.3-hr ^{132}I . Iodine-131 will be the controlling isotope with respect to the degree of retention required.¹⁴ The decay of trapped ^{131}I (and of ^{132}I , which decays to stable ^{132}Xe)

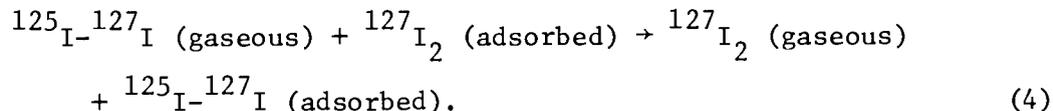
will mitigate the iodine retention problem since ^{131}I decays to stable ^{131}Xe either directly or via 11.8-day $^{131\text{m}}\text{Xe}$, which is in low yield and considerably less hazardous than ^{131}I .^{15,16} As has been indicated earlier, the DFs calculated in this investigation do not reflect decontamination resulting from decay; that is, the extent of decontamination attributable to ^{125}I or ^{130}I decay was eliminated in each case by making appropriate correction. The DFs that are to be presented for the ^{125}I -labeled I_2 are for exposure times of ≥ 35 days (both the cumulative and the incremental values being considered). Thus, in the extension of these values to apply to ^{131}I , the imposition of allowances for ^{131}I decay would result in substantially larger values. With regard to the DFs for the ^{130}I -labeled CH_3I , the time periods pertaining were short, (viz., ~ 17 hr); consequently, imposition of ^{131}I -decay allowances would result in only minor increases in these values.

4.2 Exchange Effects

Many of the DF values that were calculated in this investigation probably reflect a decontamination enhancement effect resulting from some combination of the various conceivable exchange processes such as the following:



and



Also, in cases where CH_3I was involved and the test conditions included passage through Hopcalite, equations analogous to Eqs. (3) and (4) could be written for $^{130}\text{I}-^{127}\text{I}$. Reactions similar to those represented

by Eqs. (1) and (2) are now well-established for iodized charcoal (KI-I₂ impregnated) but, of course, may not necessarily apply for silver zeolite or GX135 containing trapped iodine. The reaction of Eq. (3) is merely hypothesized, while the process of Eq. (4) appears to be highly tenable.

The DF values that may be thus affected include those which were obtained with ¹³⁰I-labeled CH₃I and for which the corresponding test bed sorbents had previously been exposed to simulated off-gas containing the ¹²⁵I-labeled I₂; also included are the I₂ DF values for cases in which the specific activity of the more recently injected I₂ differed from that previously injected. Some indication of the extent to which exchange processes may have affected certain of the I₂ results is presented in conjunction with the subsequent discussion of these and the other I₂ results for individual test situations (Sect. 4.3). A brief discussion relative to the methyl iodide results and exchange effects follows.

The CH₃I DF results corresponding to zero exposure time, except for those associated with iodized charcoal, were not exchange-affected. The other CH₃I DFs conceivably were affected to some extent, depending at least partly on the iodine loading that resulted from exposure to the simulated off-gas. For example, using the available information (estimated I₂ concentrations for different exposure periods and the value ~ 0.1 mg of I per cm³ of sorbent per CH₃I injection) and assuming one instantaneous exchange between the CH₃¹³⁰I or ¹³⁰I-¹²⁷I and all of the trapped I₂ on the test bed sorbents and on the Hopcalite bed where it pertains, calculated exchange-ascribable decontamination-enhancement factors of ~ 70 were obtained relative to the results for 286 days exposure. The analogous factors for shorter times would, of course, be less. (The CH₃I DF values, as presented, do not include any adjustments for this aspect.) For several test situations, the observed CH₃I DFs were all $\geq 9 \times 10^3$; for these situations, even the imposition of a correction factor as high as 70 would fail to reduce the DFs below 1×10^2 . Exchange effects would also be encountered, albeit in a considerably more complex manner, in an actual LMFBR fuel reprocessing off-gas system (conceivably, e.g., CH₃¹³¹I + trapped ¹²⁹I → CH₃¹²⁹I + trapped ¹³¹I, the reverse, etc.).

4.3 Observed Decontamination Factors for Iodine and Methyl Iodide

The results for the sorbents initially installed and exposed for ≤ 196 days are presented in Figs. 2-4, where DFs are plotted vs accumulated exposure time. In instances where the calculated DFs were substantially in excess of 1×10^4 , the corresponding data points have arrows pointing upward; in cases where incomplete trapping in the collection beds was indicated, the data points have arrows pointing downward. The test results for the other sorbents are given in Tables 2-6.

The performance observed for the type of iodized charcoal that was investigated (Fig. 2) indicates that its useful service life under the conditions employed in this investigation was rather limited. The data show that trapping and/or retention of iodine introduced as I_2 was adversely affected by the use of Hopcalite. As may be noted, the DFs for iodine injected as CH_3I in the case where Hopcalite was upstream remained high, presumably due to conversion of CH_3I to I_2 ; however, this good performance was more than offset by the attendant severe loss in trapping efficiency for the iodine injected as I_2 . Actually, the use of charcoal in off-gas systems where the off-gas contains oxides of nitrogen is to be avoided because of the danger of combustion, at least at higher concentrations.¹⁷ Iodized charcoal was included in this investigation for purposes of comparison with previous experience.

The results for 1/8-in. pellets of 88% silver zeolite (Figs. 3 and 4) are indicative of only fair performance, considering that DFs well in excess of 10^2 are desired. Again, the use of Hopcalite is observed to be detrimental with regard to I_2 trapping, although the results obtained with this sorbent at $100^\circ C$ are not conclusive. That the performance of this material was not better is ascribed to its large pellet size.

Satisfactory performance was observed for 1/16-in. pellets of 26% silver zeolite at $200^\circ C$ (Table 2). The DFs for CH_3I are all high, while those for I_2 are at least within the required range. It is quite probable that the actual values of those I_2 DFs preceded by a "greater than" sign were substantially in excess of the numerical values quoted. As may be noted, the I_2 DFs for 286 days exposure are considerably higher than the indicated minimums for the preceding values corresponding to 140 and 196 days. Very likely, a portion of the divergences is ascribable

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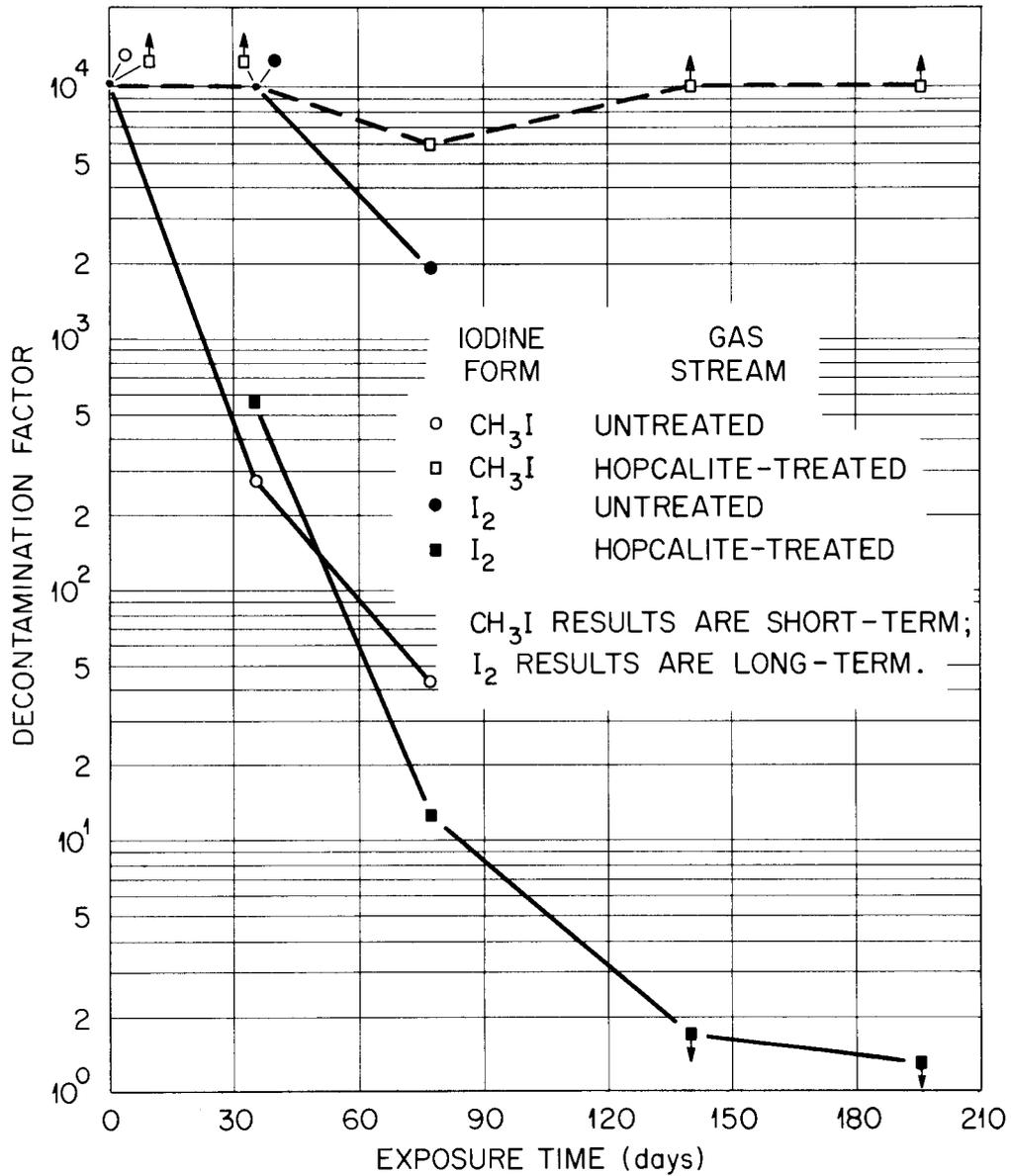


Fig. 2. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 8 x 16 Mesh (U.S.) Granules of Grade 42 Iodized Charcoal (from Witco Chemical) at ~ 66°C.

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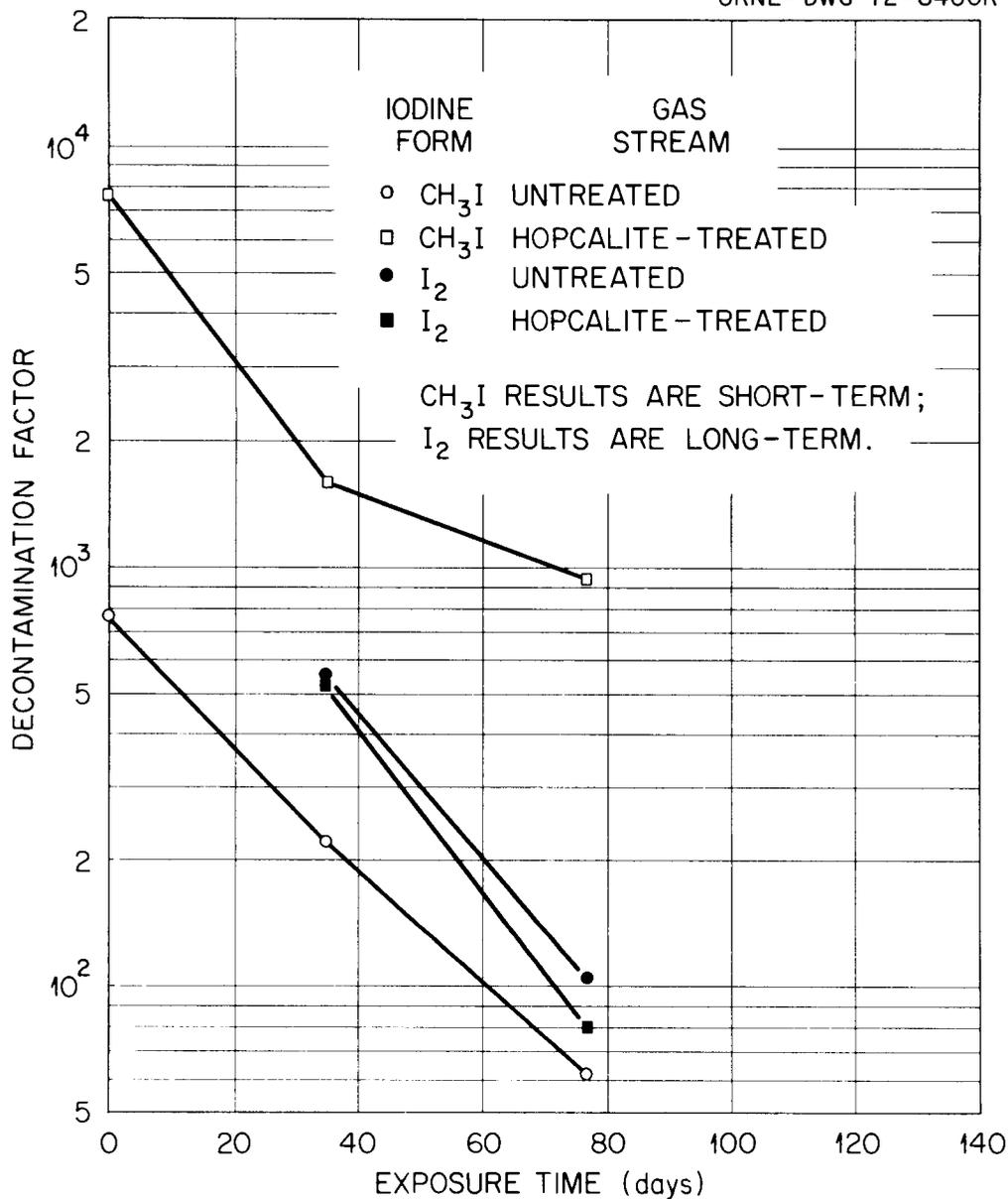


Fig. 3. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 1/8-in. Pellets of 88% Silver Zeolite (from Union Carbide) at $\sim 100^{\circ}\text{C}$.

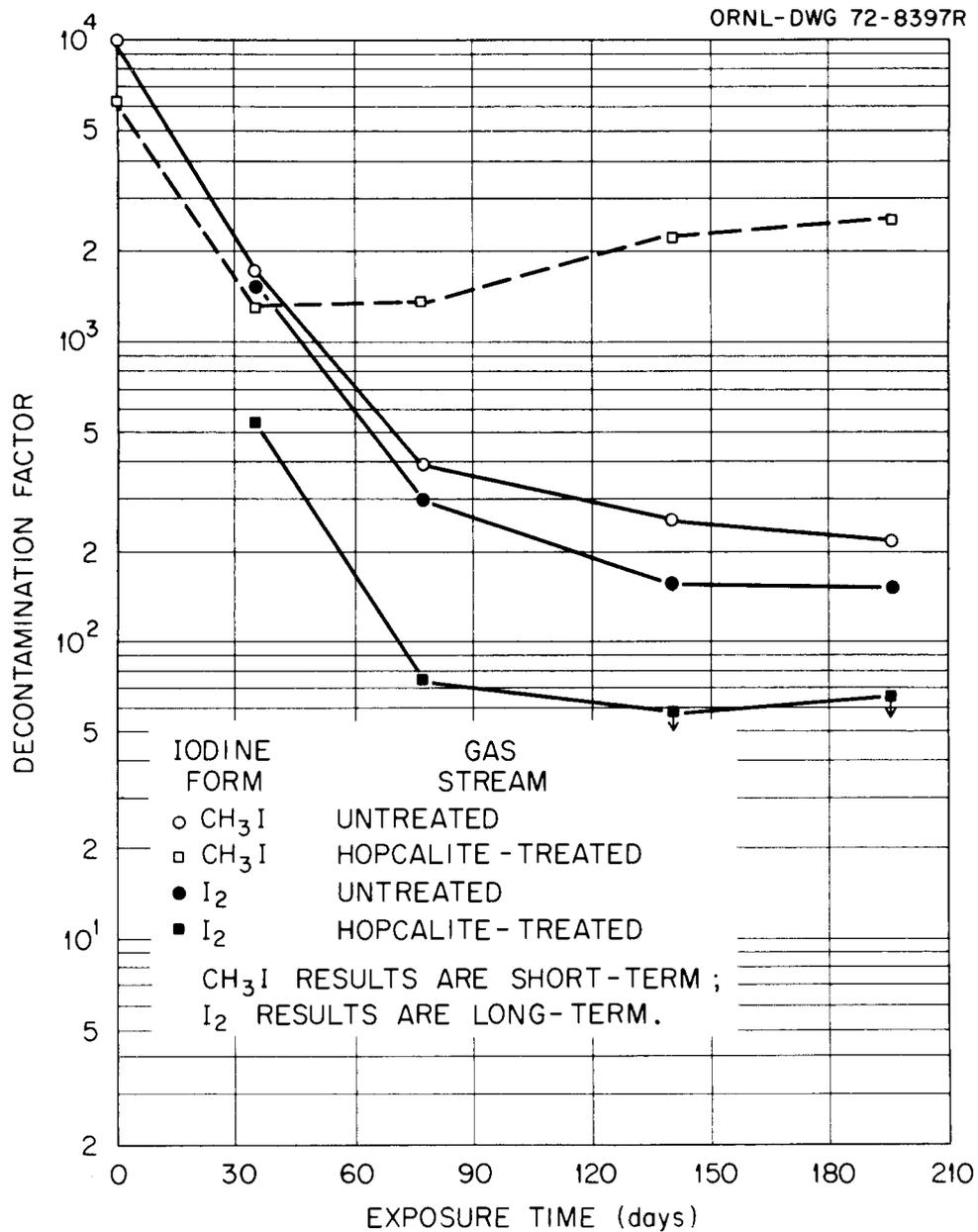


Fig. 4. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 1/8-in. Pellets of 88% Silver Zeolite (from Union Carbide) at $\sim 200^{\circ}\text{C}$.

Table 2. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 26% Silver Zeolite^a at $\sim 200^\circ\text{C}$ ^b

Cumulative Exposure Time (days)	Radioiodine Decontamination Factor (DF)			
	Without Hopcalite Upstream		With Hopcalite Upstream	
	CH ₃ I	I ₂	CH ₃ I	I ₂
0	$> 1 \times 10^4$	-	$> 1 \times 10^4$	-
35	$> 1 \times 10^4$	1×10^4	$> 1 \times 10^4$	$> 1 \times 10^4$
77	1×10^4	5×10^3	$> 1 \times 10^4$	$> 4 \times 10^3$
140	$> 1 \times 10^4$	$> 1 \times 10^3$	$> 1 \times 10^4$	$> 2 \times 10^3$
196	$> 1 \times 10^4$	$> 7 \times 10^2$	$> 1 \times 10^4$	$> 1 \times 10^3$
286	$> 1 \times 10^4$	4×10^4	$> 1 \times 10^4$	1×10^4
350	-	9×10^3	-	2×10^3

^aPrepared in the laboratory. Tests were made with 1/16-in. pellets.

^bThe DFs for CH₃I are incremental. The I₂ DFs corresponding to 286 and 350 days exposure are also incremental; however, the other I₂ DFs are cumulative.

Table 3. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of GX135^a at $\sim 200^\circ\text{C}$ ^b

Cumulative Exposure Time (days)	Radioiodine Decontamination Factor (DF)			
	Without Hopcalite Upstream		With Hopcalite Upstream	
	CH ₃ I	I ₂	CH ₃ I	I ₂
0	$> 1 \times 10^4$	-	$> 1 \times 10^4$	-
35	$> 1 \times 10^4$	$> 1 \times 10^4$	$> 1 \times 10^4$	$> 1 \times 10^4$
77	1×10^4	$> 3 \times 10^3$	9×10^3	$> 1 \times 10^4$
140	$< 4 \times 10^3$	$> 1 \times 10^3$	$> 1 \times 10^4$	$> 3 \times 10^3$
196	$< 3 \times 10^3$	$> 7 \times 10^2$	$> 1 \times 10^4$	$> 2 \times 10^3$
286	1.4×10^3	2×10^4	$> 1 \times 10^4$	$> 1 \times 10^5$
350	-	5×10^2	-	$> 1 \times 10^4$

^aFrom North American Carbon Inc. Tests were made with 8 x 16 mesh (U.S.) granules.

^bThe DFs for CH₃I are incremental. The I₂ DFs corresponding to 286 and 350 days exposure are also incremental; however, the other I₂ DFs are cumulative.

Table 4. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of GX135^a at $\sim 100^\circ\text{C}$ ^b

Cumulative Exposure Time (days)	Radioiodine Decontamination Factor (DF)			
	Without Hopcalite Upstream		With Hopcalite Upstream	
	CH ₃ I	I ₂	CH ₃ I	I ₂
63	$> 1 \times 10^4$	$> 8 \times 10^2$		
90			1×10^4	2×10^4
119	$> 1 \times 10^4$	$> 5 \times 10^2$		
154			-	$> 1 \times 10^4$
209	$> 1 \times 10^4$	$> 8 \times 10^4$		
273	-	$> 1 \times 10^4$		

^aFrom North American Carbon, Inc. Tests were made with 8 x 16 mesh (U.S.) granules.

^bThe DFs for CH₃I are incremental. The I₂ DFs corresponding to 90, 154, 209, and 273 days exposure are also incremental; however, the other I₂ DFs are cumulative.

Table 5. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 99% Silver Zeolite^a at $\sim 100^\circ\text{C}$ ^b

Cumulative Exposure Time (days)	Radioiodine Decontamination Factor (DF)			
	Without Hopcalite Upstream		With Hopcalite Upstream	
	CH ₃ I	I ₂	CH ₃ I	I ₂
49	$> 1 \times 10^4$	$> 5 \times 10^2$	$> 1 \times 10^4$	$> 1 \times 10^3$
105	$> 1 \times 10^4$	$> 6 \times 10^2$	$> 1 \times 10^4$	$> 9 \times 10^2$
195	1×10^4	$> 8 \times 10^4$	$> 1 \times 10^4$	$> 8 \times 10^4$
259	-	$> 1 \times 10^4$	-	$> 1 \times 10^4$

^aFrom Union Carbide Corporation. Tests were made with 1/16-in. pellets.

^bThe DFs for CH₃I are incremental. The I₂ DFs for 195 and 259 days exposure are also incremental; however, the other I₂ DFs are cumulative.

Table 6. Effect of Exposure to Simulated LMFBR Fuel Reprocessing Off-Gas on Radioiodine Trapping Capability of 88% Silver Zeolite^a at $\sim 200^\circ\text{C}$ ^b

Cumulative Exposure Time (days)	Radioiodine Decontamination Factor (DF)			
	Without Hopcalite Upstream		With Hopcalite Upstream	
	CH ₃ I	I ₂	CH ₃ I	I ₂
90	$> 1 \times 10^4$	$> 1 \times 10^5$	$> 1 \times 10^4$	$> 1 \times 10^5$
154	-	$> 1 \times 10^4$	-	$> 1 \times 10^4$

^aFrom W. R. Grace and Company. Tests were made with 10 x 20 mesh (U.S.) beads.

^bThe DFs for both CH₃I and I₂ are incremental.

to the limited resolution inherent in the 140- and 196-day values. Also, as already inferred, the specific activity of the iodine injected during the operating period from 196 to 286 days was substantially higher, actually by a factor of ~ 500 , than the specific activity of that which had been trapped previously. (The factor ~ 500 results from the earlier I_2 having the lower initial specific activity, plus its decay over the lengthy time span which included a certain amount of downtime.) Thus, the degree of exchange between the injected iodine and the previously trapped iodine may have been such as to have produced significantly enhanced decontamination of the former (over and above that produced by trapping and retention per se). Based on the estimated iodine concentrations, exposure times, and specific activities involved*, a calculated maximum for the enhancement factor, corresponding to the 286-day I_2 DFs and ascribable to exchange, is ~ 4 . The analogous factor for the 350-day DFs is ~ 2 . (The values in the tables do not include allowance for this aspect.) That the I_2 DFs for 286 and 350 days with Hopcalite not upstream were somewhat higher than their counterparts with Hopcalite upstream is suggestive of the behavior observed for iodized charcoal and for 1/8-in. pellets of 88% silver zeolite with respect to I_2 trapping. Possibly, the emergence of this effect for the 26% silver zeolite was a consequence of the normal decrease in trapping efficiency with higher iodine loading.

Satisfactory performance was observed for GX135 at 200°C (Table 3) for the case where Hopcalite was present upstream, and at 100°C (Table 4) without and with Hopcalite upstream. However, only a few data were obtained for the latter case. (The test bed section downstream of the Hopcalite was installed after its counterpart had been exposed for 119 days.) With GX135 at 200°C without Hopcalite upstream, satisfactory performance was observed over the first 77 days exposure. However, the 140-day and succeeding CH_3I DFs are indicative of significant deterioration in CH_3I decontamination capability; and, ultimately, an

*In the calculation of the enhancement factors relative to the I_2 DFs, allowance was made for the gradual change in specific activity of the trapped iodine resulting from the gradual addition of the more recently injected iodine to that already present.

appreciable loss in trapping capability for the iodine injected as I_2 was also indicated (referring to the 350-day value). The more definitive CH_3I DF value at 286 days, relative to the two preceding, was a consequence of changing the collection bed sorbent from iodized charcoal to 93% AgX (1/16-in. pellets). The calculated maximum exchange-ascribable enhancement factors for the 286- and 350-day I_2 DFs in Table 3 are the same as those for the analogous DFs in Table 2, but the actual factors would very likely be different for different sorbents (and, also, for different temperatures). These factors, as calculated for the similarly involved I_2 DFs in Table 4, are ~ 0.9 (i.e., < 1.0 or not really an enhancement factor), ~ 2 , and ~ 1.3 for the 154-, 209-, and 273-day values, respectively.

Satisfactory performance was exhibited for both cases by 1/16-in. pellets of 99% silver zeolite at $100^\circ C$ (Table 5) and by 10 x 20 mesh beads of 88% silver zeolite at $200^\circ C$ (Table 6), although the results for the latter are somewhat limited with respect to number and exposure time. The calculated maximum exchange-ascribable enhancement factors for the 195- and 259-day I_2 DFs associated with the 99% silver zeolite (Table 5) are ~ 2 and ~ 1.3 , respectively. This factor for the 154-day DFs associated with the 88% silver zeolite (Table 6) is ~ 0.9 .

4.4 Trapping Behavior of Elemental Iodine and Methyl Iodide

Perhaps the most striking feature of the results was the observation that, in certain of the test situations, the trapping performance for iodine injected as I_2 was adversely affected by the use of Hopcalite. In connection with this feature, the observed distribution of ^{125}I in the iodized charcoal collection beds is of interest (referring to those instances where the quantity of ^{125}I collected was appreciable). For the cases where Hopcalite was not upstream, the ^{125}I was trapped effectively in the first of the three collection beds (i.e., like I_2). When Hopcalite was upstream, significant amounts of ^{125}I were also found in the second and third collection beds (of iodized charcoal). In some of these instances, the successive amounts corresponding to a given set of collection beds decreased in a regular manner as is typical of CH_3I trapping by iodized charcoal at a relative humidity of, say, 80%. (The reasonable assumption that the distribution is described by a geometric series is employed in calculating and correcting for the radioactivity which penetrates the final collection bed.) In some other of these

cases, as denoted on the graphs by solid squares with arrows pointing downward, the ^{125}I distribution throughout the three beds (of a given set) was roughly uniform and thereby indicative of inefficient trapping.

The behavior observed for the ^{130}I -labeled CH_3I was just the reverse of that observed for the ^{125}I -labeled I_2 and was in accord with expectation. If Hopcalite was not upstream and an appreciable amount of ^{130}I penetrated to the collection beds, the ^{130}I was, in general, distributed throughout the collection beds in the descending contour characteristic of the isotopic exchange decontamination process; in two cases, which are denoted in Table 3 by "less than" signs preceding the numerical part of the CH_3I DF values, the distribution was roughly uniform. With Hopcalite upstream, the ^{130}I that penetrated a sorbent being tested was retained by the first collection bed, presumably because of the conversion of the CH_3I to I_2 .

Accordingly, the preceding observations indicate that formation of one or more organic compounds of iodine is involved in the poorer performance exhibited by certain of the sorbents with respect to trapping iodine injected as I_2 when the simulated off-gas had traversed the Hopcalite bed. This was investigated by passing portions of the two gas streams, one treated and the other untreated (with Hopcalite), through iodine characterization packs of the type developed by Bennett, Hinds, and Adams.¹¹ These packs differentiate between reactive iodine (I_2), iodine associated with airborne particulate matter, and organic iodides. The sampling ports were located near the inlets to those test bed sections operated at 66 or 100°C. At least 98% of the ^{125}I -labeled iodine in either stream was indicated to be in the elemental form. These data, together with the related observations, are consistent with the following suggested possibilities: (1) that organic fragments in the Hopcalite-treated stream react with iodine trapped on the test beds to yield organic iodides of which some fraction is evolved, and (2) that dodecane in the untreated stream undergoes little reaction with the trapped iodine.

4.5 Concerning the Desirability of Using a Catalytic Oxidation Bed

As mentioned in Sect. 2, the secondary objective of this investigation was to ascertain the desirability of having a catalytic

oxidation bed upstream of the sorbent. Actually, this concept was only investigated from a rather narrow standpoint since only one type of oxidizing catalyst (viz., Hopcalite No. 21215) and, aside from the use of different sorbent temperatures and the described variation in I_2 concentration, only one set of operating conditions were involved. In general, the CH_3I and I_2 DFs observed for the sorbents exhibiting satisfactory performance were relatively high and at or near the limit of the pertaining analytical resolution regardless of whether the gas stream had or had not traversed the Hopcalite bed. However, in the case of GX135 at 200°C (Table 3), the use of Hopcalite did appear to offer a distinct advantage. On the other hand, certain of the I_2 DFs for 26% AgX (Table 2) were suggestive of a detrimental effect. Results for the less effective sorbents (Figs. 2-4) were generally indicative of a large beneficial effect (due to Hopcalite) relative to the trapping of iodine introduced as CH_3I but were indicative of a small to large detrimental effect relative to the retention of iodine introduced as I_2 . Since the detrimental effect appears to be associated with the formation of organic iodides which, in turn, may have resulted from incomplete conversion of dodecane to CO_2 and H_2O , the use of a Hopcalite bed that provides a longer residence time conceivably would result in mitigation of the detrimental effect. However, the possible efficacy of this approach was not investigated.

Thus, the results obtained for both the satisfactorily performing sorbents and the less effective sorbents were generally inconclusive regarding the value of pretreating the off-gas. On the other hand, the results for the satisfactorily performing sorbents, taken as a whole, do tend to indicate that the possible benefit to be gained by use of a catalytic oxidation bed in the off-gas system would probably not be of sufficient magnitude to warrant the associated added cost.

4.6 Sorbent Shape and Particle Size

Pence, Duce, and Maeck² have pointed out that granular sorbents present a greater apparent surface area than those in the form of pellets or beads and, consequently, would provide the higher trapping efficiency; they cite an example in which 12-14 mesh AgX granules exhibited a DF higher than that obtained for 14-18 mesh AgX beads

by a factor of nearly 20 despite the smaller particle size of the latter. Thus, even better performance than indicated by the results herein for 1/16-in. silver-sodium zeolite pellets should be attainable with otherwise similar material in a granular form. The 1/16-in. dimension corresponds to a mesh No. between 12 and 14 (U.S.); therefore, the use of silver-sodium zeolite granules with a mesh size range of, say, 10 x 16 would appear to be appropriate.

The mesh size range 8 x 16 (U.S.) is usually specified for sorbents employed in nuclear applications;¹⁸ the use of finer material than that retained by a No. 16 sieve is presently limited by the mechanical difficulties associated with supporting and retaining it. (The above-mentioned 10 x 16 range would not be expected to represent an application problem.) Accordingly, in this work, the emphasis was placed on sorbents which were 8 x 16 mesh or roughly the equivalent in size (viz., 1/16-in. pellets). Of course, all else being equal, the sorbent with the smaller particle size is the more effective, and, if the application difficulties can be overcome, consideration should be given to the application of a finer sorbent than that represented by the mesh size ranges 8 x 16 or 10 x 16.

5. ACKNOWLEDGMENTS

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