

DATE ISSUED **APR 10 1979**

ORNL/TM-6607

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A Literature Survey of Methods to Remove Iodine from Off-Gas Streams Using Solid Sorbents

R. T. Jubin



OAK RIDGE NATIONAL LABORATORY
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Printed in the United States of America. Available from
the Department of Energy,
Technical Information Center
P.O. Box 62, Oak Ridge, Tennessee 37830
Price: Printed Copy \$6.00; Microfiche \$3.00

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ORNL/TM-6607
Dist. Category UC-79c

Contract No. W-7405-eng-26

CONSOLIDATED FUEL REPROCESSING PROGRAM

A LITERATURE SURVEY OF METHODS TO REMOVE
IODINE FROM OFF-GAS STREAMS USING SOLID SORBENTS

R. T. Jubin⁰³

Chemical Technology Division

Date Published: March 1979

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HIGHLIGHTS

During the past two decades, various systems have been studied to reduce the iodine released to the environment. Two liquid systems are available: the Iodex system and the mercuric nitrate-nitric acid (Mercurex) system. Solid sorbents have also been studied, either as secondary systems to provide final filtering following use of either the Iodex or Mercurex system or as primary systems to replace the liquid methods. These solid sorbents include silver zeolite, silver mordenite, alumina silicates, and macroreticular resins. Activated carbon has also been examined as a reference material. Activated carbon, however, cannot be considered as a primary sorbent for reprocessing plant off-gas, because of its low ignition temperature and its adverse reactions with nitrogen oxide which could lead to the formation of explosive compounds and to poisoning by organic contaminant in the off-gas.

The most promising system to date for primary iodine removal is the Iodex system. This system has already been tested on an engineering scale and has demonstrated good capabilities and operability. There are, however, concerns about the long-term integrity of the piping in contact with the 20 to 23 M HNO₃. Silver mordenite appears to be the most promising of the solid sorbents developed thus far. This material will, however, require additional testing with CH₃I, in addition to engineering-scale tests of filter beds and regeneration facilities. A silver-containing alumina silicate, AC-6120, also shows potential on a once-through basis. Silver zeolites are capable of adsorbing iodine up to 500°C. Silver zeolites, like most other sorbents, are adversely affected by water vapor especially when condensation occurs. The high cost of silver and its limited regeneration potential, plus the lack of acid resistance of the zeolite, makes it less attractive than silver mordenite or AC-6120 for large-scale use.



1. INTRODUCTION

Iodine-129 is produced by both natural and man-made sources. The natural sources include the spontaneous fission of uranium and the interaction of cosmic rays with xenon in the atmosphere. The annual production through these natural paths is approximately 10 mg/year. With its long half-life, 1.6×10^7 years, the estimated amount of ^{129}I was 2×10^5 g as of 1940.¹

The dawn of the nuclear age brought with it a new source of radioactive iodine. A single light water reactor (LWR) will produce 234 g of ^{129}I based on 30,000 MWd/t of uranium burnup.² In handling the spent fuel, a single 5-t/d nuclear fuel reprocessing plant would liberate 3.2×10^5 g of ^{129}I /year. This would result in an extremely fast buildup of ^{129}I in the biosphere if no iodine removal steps were taken.

In addition to ^{129}I , various amounts of other short half-life iodine isotopes are also produced. However, none of these appear in significant amounts after long (<200-d) decay times. On the other hand, in the reprocessing of liquid-metal fast breeder reactor (LMFBR) fuels, reducing the decay times to as little as 90 d might have economic incentive. This short decay time would mean that larger amounts of the short half-life isotope ^{131}I ($t_{1/2} = 8.05$ d; specific activity, 1.24×10^5 Ci/g) would also be released.

One of the challenging problems to be solved in the nuclear industry (for short-cooled fuels) is the required removal and retention of additional amounts of iodine. This study will examine the available literature in this area, with its main emphasis being on the removal of iodine from gas streams by solid sorbents. The study will also attempt to point out areas where additional work remains to be done.

2. BACKGROUND AND IODINE REMOVAL REQUIREMENTS

The Windscale No. 1 pile accident in October 1957 pointed out the importance of the retention of fission product ^{131}I as a potential reactor safety problem. The release of ^{131}I was the result of a fire in the air-cooled reactor.³ It has been assumed that any release of fission products in an accident condition would appear as particulates, and the filter system in use was designed to contain such a release. However, the 1957 accident showed that this was not the case when only about 50% of the iodine was retained by the filter system.⁴

Since 1957, a great deal of research has been directed toward the removal of iodine from gas streams. The initial studies were devoted to the retention of iodine within a reactor confinement building in a post-accident situation. More recently, the main thrust of the research has been directed toward the removal of iodine from the off-gas streams of nuclear fuel reprocessing plants.

The release limits presented in this section are based on historical information; consequently, the design engineer must use the most recent regulations available to determine current limits.

Fuel reprocessing facilities are required to have off-gas systems designed "to confine hazardous chemicals or radioactive materials evolved during process operations and radioactive waste storage, and to reduce their concentrations in gaseous process effluents to levels as low as practicable."⁵ In addition, all parts of the off-gas system should be designed to meet the release limits for radioactive materials during normal operation. These limits are set forth in regulation 10 CFR 20, which leaves some leeway on the permissible concentration of radioactive material discharged from the stack, providing the discharge site is within a restricted area. In the case of a restricted area, the amount discharged may be calculated from the limit set forth in Table II of Appendix B of 10 CFR 20 by applying the proper factors for dilution, decay, and dispersion between the point of discharge and the boundary of the restricted area.

In the late 1960's and early 1970's, a number of reports were published concerning the maximum permissible concentration (MPC) of iodine that could be released safely to the environment.⁶⁻⁹ The amount of iodine released must take into account the biological pathways, the concentration of isotopes along these pathways to the human body, and the concentration effects in the body itself (e.g., for iodine, 20% reports to the thyroid).

Bryant⁹ determined "working or controlled release limits" for ^{131}I found in milk to be 400 pCi/liter, and an "emergency release limit" for deposition on pasture land to be $1.5 \mu\text{Ci}/\text{m}^2$. Burnett¹⁰ explains the "factor of 700," which applies to the concentration effects in the air-grass-cow-milk-child pathway for ^{131}I . This factor is based on the correlation between ^{131}I deposited on the grass and the ^{131}I concentration in the milk from cows grazing on this grass, that is, $1 \mu\text{Ci}$ of ^{131}I per square meter of grass = $0.09 \mu\text{Ci}$ of ^{131}I per liter of milk. This relationship was established after the Windscale incident. Using this relationship, the effective half-life of 5-d ^{131}I on grass coupled with the Federal Radiation Council's recommended dose limit of 80 pCi of ^{131}I per day shows that the legal environmental release limit for ^{131}I of $1 \times 10^{-10} \mu\text{Ci}/\text{cm}^3$ is a factor of 700 too high. However, it must be kept in mind that this factor is no more than a "rule of thumb," since it does not take into account any localized meteorological factors or the details of the critical paths that the isotope will follow.

Tadmor⁸ points out the need for ^{129}I control. Based on the permissible thyroid burden of 6×10^{-8} Ci (Ref. 11), it appears that the maximum amount of ^{129}I in the thyroid should be 5.4% of the total iodine content. From this, assuming an average concentration of stable iodine in the air of $10^{-7} \text{g}/\text{m}^3$, the corresponding airborne activity should be $8 \times 6 \times 10^{-13} \text{Ci}/\text{m}^3$, which is a factor of 23 less than the legal limit for unrestricted areas of $2 \times 10^{-11} \text{Ci}/\text{m}^3$ (presented in 10 CFR 20).¹² The required release limits for reprocessing LMFBR fuels have been assessed by ORNL's Chemical Technology Division.¹³ The requirements are based on limits of 4.8×10^{-14} and $1 \times 10^{-15} \text{Ci}/\text{m}^3$, respectively, for ^{131}I and ^{129}I at 2 to 3 km from the site. Also presented were the projected decontamination factors (DF's) for various type plants using these selected release limits. The

DF factors were derived on the basis of computer codes which took into account the various complex factors of plant siting and the numerous biological pathways.

Federal regulation 40 CFR 190, revised as of July 1, 1977,¹⁴ limits the public dose from a discharge of radioactive materials (except for radon and its daughters) to less than 25 millirems/year to the whole body, 75 millirems/year to the thyroid, and 25 millirems/year to any other organ. In addition, the total quantity of ^{129}I that may be released to the general environment from the fuel cycle must be less than 5 mCi/GW-year.

Figure 1 (from Ref. 15) is a typical flowsheet for off-gas control. It is expected that the major portion of the iodine will be driven off early in reprocessing into a relatively small volume of gas. The question remaining before the nuclear industry is what method should be used to remove and retain the iodine evolved during reprocessing.

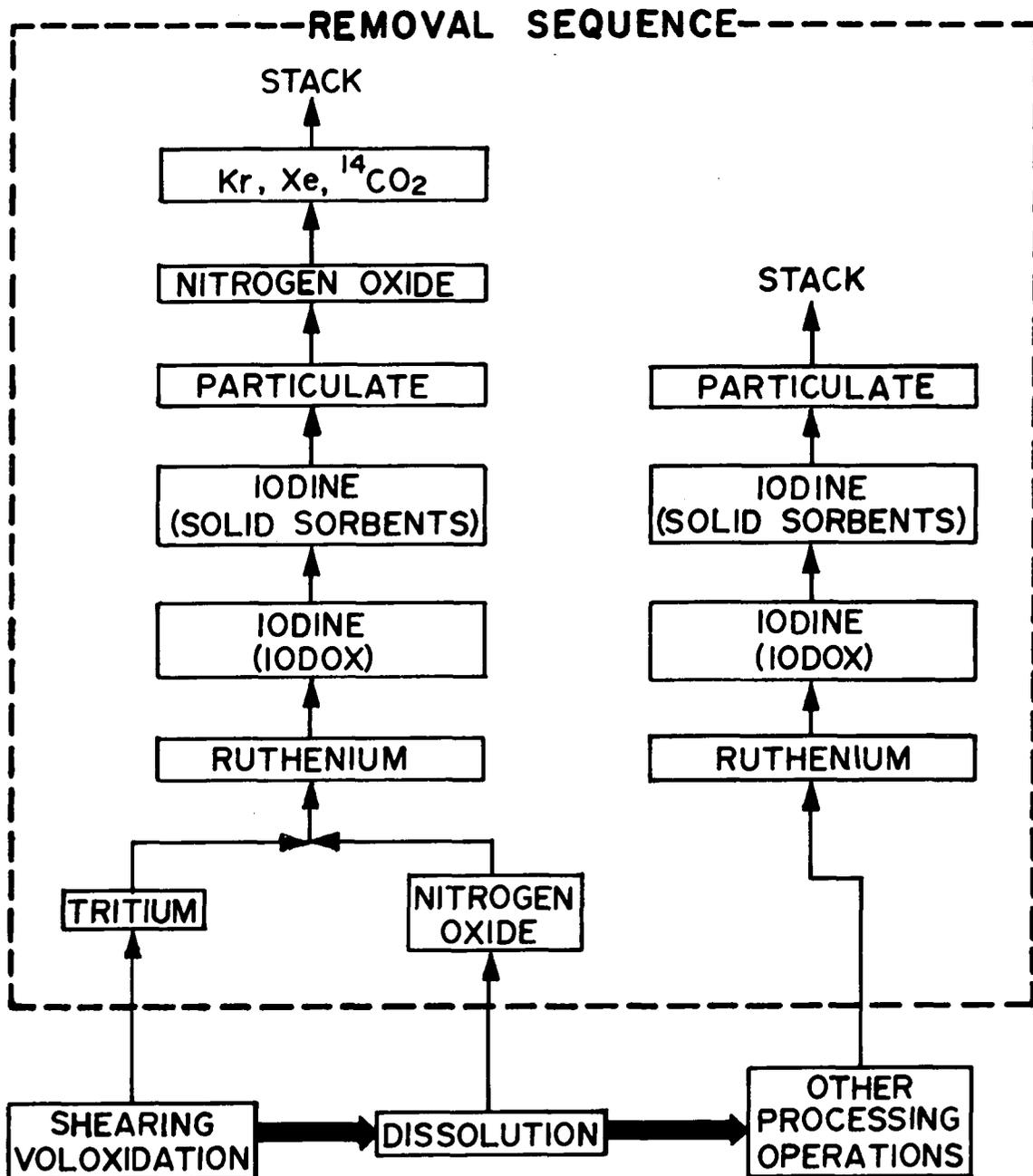


Fig. 1. Off-gas treatment flowsheet for a fuel reprocessing plant.

3. IODINE CHEMISTRY

3.1 Iodine Forms Expected

Table 1 (from Ref. 16) shows the various fission product iodine isotopes, their specific activities, and the amounts expected from a typical reactor fuel. Those of most concern to the nuclear fuel reprocessing industry include:

- (1) Iodine-131 ($t_{1/2} = 8.04$ d) is the most important from the standpoint of safety and also in determining radiation exposure limits to power station crews during normal and accident conditions. This isotope is also noteworthy in the reprocessing of short-cooled fuel because of its high specific activity.
- (2) Iodine-133 ($t_{1/2} = 21$ h) must be considered in calculating radiation exposure in a power station. The uptake by inhalation is of practical importance.
- (3) Iodine-132 ($t_{1/2} = 2.3$ h), Iodine-134 ($t_{1/2} = 54$ min), and Iodine-135 (6.6 h) are only of minor importance because of their short half-life. However, because of the decay heat in the unlikely event of an accident, these isotopes must be considered in the design calculations for iodine filters in power stations. Most of the iodine inventory is volatilized in the filters.
- (4) Iodine-129 ($t = 1.7 \times 10^7$ years) and Iodine-127 (stable) make up the major fraction ($\sim 87\%$)¹⁶ of the fission product iodine. The bulk of these isotopes must be taken into account when designing filters for both power stations and reprocessing plants. One of the factors that make Iodine-129 a potential hazard is that it is a low beta and gamma energy emitter, which makes detection and measurement difficult.

Since iodine is chemically very reactive, it must be assumed that when iodine is released in its elemental form, other compounds will be formed when suitable reaction partners are present.

Table 1. Typical fission product iodine content of a typical 20-MW(t) reactor at shutdown

Iodine isotope	Half-life (t 1/2)	Specific activity (ci/g)	Core inventory at shutdown for 20-MW(t) reactor operated (Ci) 300 d	Total iodine mass at shutdown (mg)
127	Stable			9,720
128	25.0 M	5.87×10^7	1.80×10^2	~0
129	1.7×10^7 years	1.7×10^{-4}	5.61×10^{-3}	32,500
130	12.4 h	2.0×10^6	3.03×10^3	2
131	8.05 d	1.24×10^5	5.64×10^5	4,560
132	2.3 h	1.03×10^7	7.96×10^5	77
133	21.0 h	1.12×10^6	1.02×10^6	904
134	53.0 min	2.65×10^7	1.28×10^6	48
135	6.7 h	3.47×10^6	1.08×10^6	313
136	83.0 s	1.0×10^9	4.61×10^5	~0
137	23.0 s	$>10^9$	8.39×10^5	~0
138	5.9 s	$>10^9$	5.92×10^5	~0
139	7.0 s	$>10^9$	2.97×10^5	~0
Totals			7.928×10^6 Ci	48,126 mg

3.2 Methyl Iodide

The simplest organic iodine compound is methyl iodide (CH_3I), which is formed by the reaction of iodine and small quantities of methane (4.5×10^{-8} mole/liter)¹⁷ always present in the air. Methyl iodide was identified as early as 1963¹⁸ during tests in room air following the release of elemental iodine. Subsequently, this compound was also identified in the room and exhaust air and in the off-gases from reactors. A summary of the physical properties of CH_3I may be found in Appendix A. Higher alkyl iodides arising from reactions with oil, grease, solvents, and paints found within the test facility have also been noted.

L. F. Parsly¹⁷ has prepared an in-depth summary of the chemical and physical properties of CH_3I . Parsly has examined the work of Golden and Walsh, and Benson, Goy, and Pritchard (who published their results almost simultaneously) on the equilibrium of



The attainment of equilibrium is slow, requiring several hours. Since a major portion of the observations by other investigators occurred at temperatures greater than 400°C , they are of little significance in off-gas handling.

Parsly also examined studies of radiation effects on CH_3I . These effects fall into two major categories: photolysis and radiolysis. Extensive work dating back to 1920 has been done in the field of photolysis of alkyl halides. In the 1930's, it was recognized that



where h is Planck's constant and ν is the frequency of the light.

Ultraviolet light at 2537 Å is required to rupture the C-I bond. The yields from this process were low and were attributed to the recombination of CH_3 and I ; however, in the presence of silver, the rate was 40% faster, and all of the iodine appeared as AgI . The gaseous products were 55% C_2H_6 , 18% C_2H_4 , and 28% CH_4 . The high yield was accounted for

by assuming that the silver collected the iodine, preventing recombination. Also, when nitric oxide is present, recombination is suppressed. Carbon dioxide slows the process in the absence of silver, but has no effect when silver is in the system.

The literature to be found on radiolysis is quite extensive; however, much of the information concerns liquid-phase reactions. Tang and Castleman¹⁹⁻²¹ indicate that the rate of decomposition of CH₃I in air is a function of dose to air and the initial CH₃I concentration. Concentrations of 10⁻⁸ to 10⁻⁶ mole of CH₃I per liter of air were reduced by a factor of 10 by a dose of 5 × 10⁵ rads. It also appeared that water vapor hindered the decomposition.

3.3 Hypoiodous Acid

Iodine undergoes hydrolysis in aqueous solutions to form hypoiodous acid (HOI) according to



The existence of airborne HOI in a steam air atmosphere was first postulated in 1968 by Keller et al. of Idaho Nuclear Corporation.²² Numerous investigators²³⁻²⁵ have observed an iodine compound (neither elemental nor organic) in a humid atmosphere.

Although HOI appears to have a long half-life in air, about 1/2 d, there is no direct proof that it exists as a gaseous component or as a stable airborne species. There are, however, several reactions which indirectly indicate its presence in off-gas and exhaust air streams.

Experiments by Keller et al.²⁶ have shown that the optimum conditions for the formation of HOI are an iodine concentration in liquid of approximately 10⁻⁶ M, a pH of 10, and a temperature of 90°C (see Fig. 2). Keller also presents equilibrium data for the hydrolysis of iodine. Kabat²⁷ has also presented the results of an extensive study of the relations and kinetics for the production of HOI.

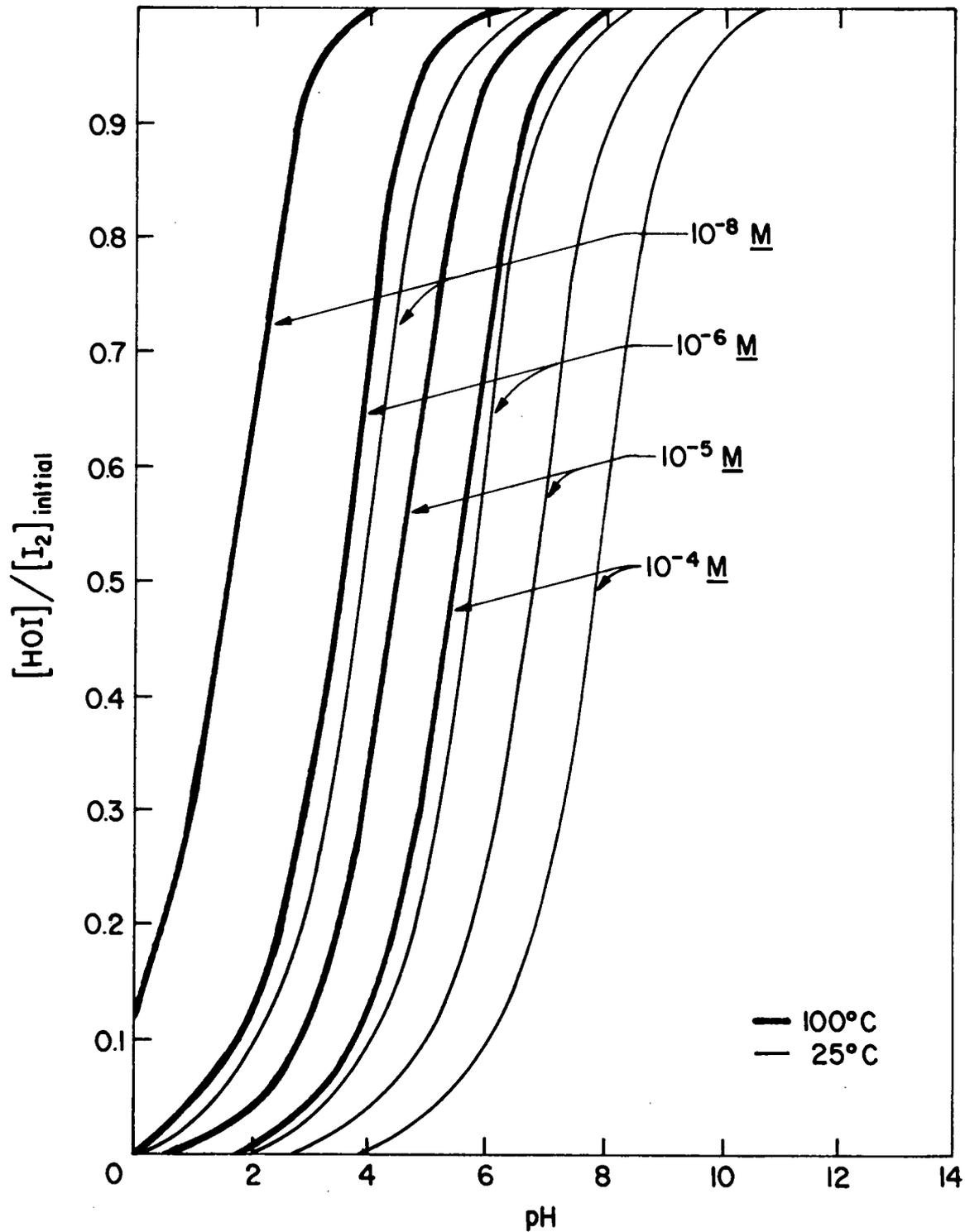


Fig. 2. Mole ratio of $(\text{HOI})/([\text{I}_2]_{\text{initial}})$ as a function of pH and initial molarity of elemental iodine (based on information from Ref. 26).

3.4 Hydrogen Iodide

Another inorganic form of iodine that must be considered is hydrogen iodide (HI), which is only expected to occur in reducing conditions.²⁸

Undoubtedly, there are many other radioiodine compounds in the vapor phase yet to be found in effluents from both power stations and reprocessing plants. In a number of reported tests, iodine compounds have been found that are difficult to separate and to chemically define.

4. WET SCRUBBING SYSTEMS

Iodine removal in the reprocessing industry has generally been classed into three broad categories: primary, secondary, and final cleanup systems. The primary iodine removal systems are designed to remove the iodine volatilized in the dissolver. The secondary system provides additional removal from the dissolver off-gas and from other process equipment. Final iodine removal is provided by an additional off-gas cleanup system before the gas is released to the stack.

Several wet scrubbing techniques have been proposed for the primary removal step. These processes employ either mercuric nitrate-nitric acid ($\text{Hg}[\text{NO}_3]_2\text{-HNO}_3$), the Mercurex process, or hyperazeotropic (>20 M) nitric acid, the Iodox process. Both of these processes are discussed in some detail in a report by Holladay.²⁹ Two mercuric nitrate units in series have been installed in the Barnwell (S.C.) Nuclear Fuel Plant; the Mercurex process is also being tested at the Belgian Nuclear Research Center. Considerable work has been accomplished by J. G. Moore et al.³⁰ on the separation, removal, and storage of iodine from the $\text{Hg}(\text{NO}_3)_2\text{-HNO}_3$ solution. Some problems associated with this process are (1) the low recovery for aromatic iodides and (2) the complexity of the separation of iodine from the $\text{Hg}(\text{NO}_3)_2\text{-HNO}_3$ solution.

The Iodox process (Fig. 3) which employs 20 to 23 M nitric acid to scrub iodine from the gas stream is under development at Oak Ridge National Laboratory.^{31,32} This process has the capability of providing decontamination factors (DF's)* greater than 10^4 .¹ The process calls for the removal of iodine as anhydrous iodic acid (HI_3O_8) from the evaporator. Since HI_3O_8 is very soluble in water,³³ it should be converted to a more insoluble form such as barium iodate [$\text{Ba}(\text{IO}_3)_2$] before final storage; this process is also being studied by Moore.³⁰ The Iodox process has the potential advantages of (1) handling a high NO_2 concentration of 7.6% and still provide DF greater than 10^5 (Ref. 34) and (2) accepting a humid off-gas stream with only a decrease in efficiencies on the first

* See Appendix B for definition.

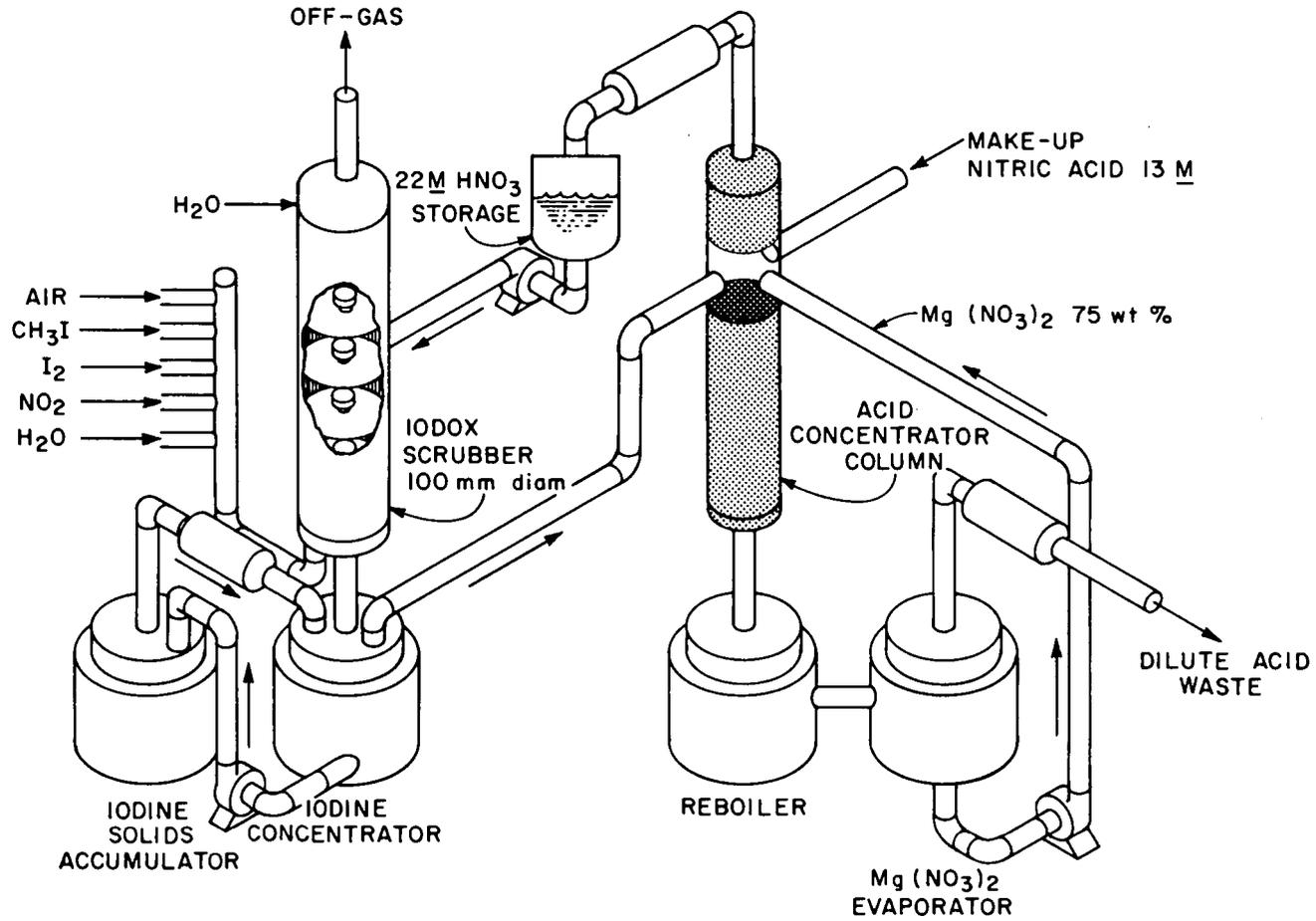


Fig. 3. Iodex process flowsheet for iodine removal.

plate. However, the equipment is expected to be more expensive than that for the mercuric nitrate process because it must be designed to handle the highly concentrated nitric acid, and more involved as the system includes not only the scrubber tower but also the extractive distillation unit and an iodine accumulator, reboiler section.

Both wet scrubbing techniques and solid sorbents have been proposed for the secondary removal system. The solid sorbents suggested, which are also proposed in most plant designs for the final iodine removal system, include charcoal, zeolites, amorphous silicic acid impregnated alumina, mordenites, and macroreticular resins.

5. SOLID IODINE SORPTION MATERIALS

In general, solid sorbents offer several advantages over the liquid systems mentioned above: (1) they allow a simpler design; (2) they are comparatively noncorrosive; (3) the waste product is in a dry form that is easy to handle and contain; and (4) the simpler design, with few moving parts, makes for higher reliability and lower maintenance costs.

5.1 Activated Carbon

Of the solid sorbents proposed for iodine removal, activated carbon has been most widely studied. Presently, nuclear power plants almost exclusively use impregnated, activated carbon for the removal of radioiodine. It was discovered that the impregnation is required to remove CH_3I from high humidity gas streams; the most widely used impregnates at present are I^- , KI , and triethylenediamine (TEDA). Although activated carbon has been used successfully in power plants, it has several serious drawbacks.

- (1) It has a relatively low ignition point; thus, without continuous air flow through the filter, the decay heat of trapped radioiodine could ignite the bed.
- (2) The bed has poor iodine retention at high temperatures and permits total iodine releases upon ignition.
- (3) The presence of nitrogen oxides adversely affects the bed's performance; nitrogen oxides can also lead to the production of explosive compounds within the filter bed.^{35,36}

These factors rule out the use of this material in reprocessing plant off-gas systems; therefore, this paper will only consider the effects of various parameters on the iodine adsorption of activated charcoal.

The removal of radioiodine from gas streams by activated carbon appears to be the function of many variables. Because of worldwide interest in this material, numerous reports and papers have been published which deal with iodine removal (see Table 2) as affected by the following parameters: inlet iodine concentration, relative humidity, bed temperature, service time and weathering, radiation, impregnation, manufacture,

Table 2. Summary of literature reference numbers pertaining to parameters affecting the removal efficiency of activated carbon

Concentration	— 4, 16, 27, 44, 45, 47, 48, 49, 53, 54, 55, 56, 57, 58
Gas velocity	— 35, 39, 40, 45, 48, 49, 51, 54, 55, 56, 57, 59, 60, 61, 62, 63, 64
Particle size	— 38, 49, 59, 64
Manufacture/ type	— 4, 37, 45, 49, 50, 54, 56, 60, 61, 63, 65, 66, 67, 68, 69, 70
Impregnation	— 27, 35, 37, 38, 39, 40, 44, 45, 49, 50, 53, 54, 55, 56, 59, 60, 61, 63, 67, 68, 69, 70, 71, 72
Bed depth	— 43, 49, 56, 57, 60, 68, 70, 71, 73
Aging	— 37, 39, 40, 41, 45, 49, 51, 56, 58, 59, 61, 62, 68, 74, 75, 76, 77
Operating temperature	— 37, 39, 41, 43, 45, 54, 56, 57, 59, 61, 62, 64, 69, 70, 77
Ignition	— 35, 38, 39, 40, 54, 56, 59, 61, 69
Relative Humidity	— 43, 44, 49, 50, 53, 56, 57, 59, 61, 62, 63, 64, 68, 70, 71
Radiation	— 37, 40, 45, 54, 61, 62, 65, 68, 77
Oxidizing Beds	— 50, 51, 75
Regeneration	— 39, 45, 62, 77

particle size, and gas velocity through the filter. Mechanisms responsible for the adsorption of the iodine onto the charcoal will also be discussed in this paper.

5.1.1 Removal of elemental iodine

Elemental iodine can be bound to activated carbon by either chemisorption, which is more preferred, or physical adsorption. Physical adsorption is the initial method of adsorption on activated carbon, which is due to its large surface area (BET areas = 800 to 1000 m²/g) and pore structure. The various functional groups, phenolic and other alcoholic hydroxyl groups, may create the chemical bonds, which greatly increase the amount of iodine retained during desorption tests, with the iodine after it is physically adsorbed.

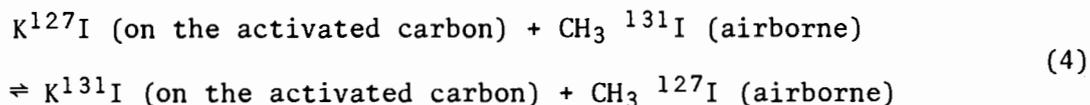
5.1.2 Removal of methyl iodide

The simultaneous adsorption of water vapor from the gas stream greatly impairs the adsorption of methyl iodide making it somewhat more difficult to remove than elemental iodine. At low relative humidities (<5%), there is little effect from the water vapor. As the humidity is raised into the intermediate range, capillary condensation occurs in the micropores, and some of the larger pores are also filled with water. When the relative humidity exceeds about 30%, the adsorption of methyl iodide (CH₃I) is negligible.

5.1.3 Use of impregnated carbon to remove CH₃I

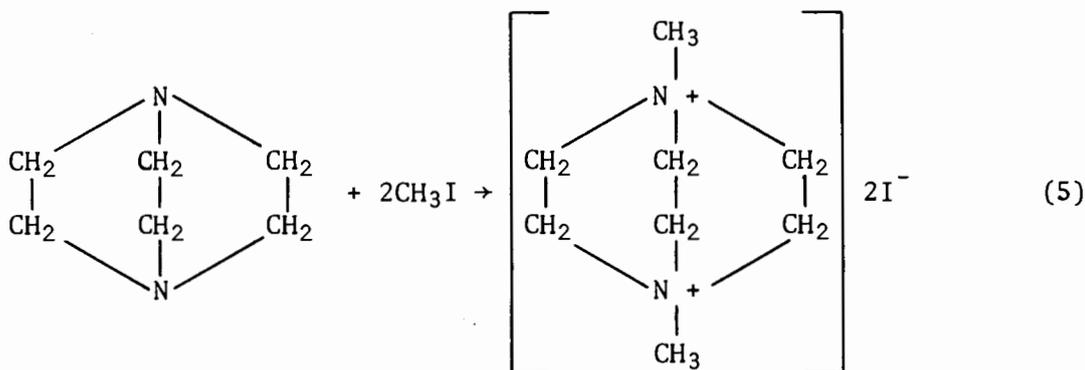
To obtain acceptable adsorption of CH₃I at a high relative humidity, activated carbon has been impregnated with numerous materials that provide locations for either chemisorption or isotopic exchange of the iodine. The three most common impregnates are K¹²⁷I, I₂, or triethylenediamine (TEDA), [1,4-diazobicyclo (2.2.2) octane], in amounts ranging from 0.5 to 5 wt%.

K¹²⁷I provides a site for isotopic exchange with the other airborne iodine isotopes according to:



For this reaction, the rate controlling steps appear to be diffusion of the active methyl iodide to the surface of the carbon and desorption of the inactive CH_3I from the surface. Large excesses of K^{127}I are required to drive the reaction far to the right.

TEDA is a white hygroscopic crystalline material which has a very narrow liquid range (MP. 159.8°C , BP. 174°C) and a low flash point on carbon (190°C). A quaternary ammonium salt bound to the carbon is formed by a chemical bond of TEDA with methyl iodide.



These reactions also apply to some degree for other organic iodine compounds.

In a scoping study for elemental iodine sorbents, Evans and Jones³⁷ reported that (1) KI_3 impregnation methods vary widely from vendor to vendor, (2) highly insoluble PbI_2 had no noticeable benefit over KI_3 , (3) carbons impregnated with TEDA perform better than the others in a radiation or NO_2 environment, and (4) TEDA + KI coimpregnated carbons performed nearly as well as those with TEDA.

Later, Evans³⁸ showed that carbon impregnated with both TEDA and KI performed better than either alone. As a result, a 10×16 mesh coconut shell carbon containing 1% TEDA and 2% KI with a flame retardant, was selected to be used in the Savannah River confinement system; it produced a removal efficiency for I_2 of 99.983% when operated at 80°C and 100% relative humidity in a radiation field of 3×10^7 rd/h.

While adsorption of CH_3I is improved by the addition of impregnates, Milham³⁹ reports that these have significantly inferior desorption properties at a high temperature compared to unimpregnated carbon and should be restricted to low temperature use.

5.1.4 Removal of hypiodous acid (HOI)

Until recently HOI was unknown, which resulted in work being quite sketchy at this point. Kabat²⁸ reported in 1974 that HOI was more difficult to remove than CH_3I and was more easily desorbed from carbon. He also reported that activated carbon impregnated with iodine salt on TEDA was a more efficient sorbent than the other tested impregnated carbons; Kabat also noted that HOI adsorption decreases with increasing relative humidity.

Although the exact mechanism for trapping HOI has not been verified, one suggested mechanism for trapping HOI is that it is physically adsorbed and then reacts with the impregnate; nevertheless, much work remains to be done in this area.

5.1.5 Effects of temperature

The retention of elemental iodine is most strongly influenced by the operating temperature of the bed, the service history of the carbon, and the radiation exposure of the bed after loading. These factors also influence the retention of organic iodides with the addition of the amount and type of impregnation in the carbon bed and the relative humidity of the gas stream.

In general, higher temperatures of the gas will result in (1) a high rate of desorption of the iodine, (2) an increased oxidation rate of the carbon, and (3) ignition of the bed itself and total desorption of the iodine.

In most cases, activated carbon made from coconut shells will show desorption rates at high temperatures lower than carbons based on wood, petroleum, or coal. This desorption behavior of activated carbon should limit the operating temperature of the filter bed to 120°C .²⁸

Numerous studies have been conducted to study the effects of ignition temperature versus impregnation. In general, due to the low flash point of TEDA, the higher the TEDA content, the lower the ignition temperature. Even with flame retardants added, the oxidation of TEDA in 2% TEDA carbons provided enough heat to overcome the flame retardants suppressive effects. There was also no significant difference in TEDA ignition temperatures of carbons made by various manufacturers.⁴⁰ Table 3 indicates the average ignition temperatures for activated carbons without flame retardants added. The presence of NO₂ in the gas stream will also lower the ignition temperature.³⁵ The gas velocity also affects the ignition temperature; by lowering the velocity, less heat is removed and the ignition temperature is lowered.³⁹

Kovach³⁷ notes that the ignition temperature is not a characteristic of the carbon itself but is a characteristic of a system consisting of the carbon, gas flow and composition, and carbon impregnates.

5.1.6 Service history

During normal use, the carbon filters come in contact with solvent vapors, oil vapors, SO₂, and NO_x which all tend to decrease the amount of iodine adsorbed and increase the iodine desorption rate. This results from the consumption of the active adsorption sites in the carbon (Fig 4).⁴¹

The following measures have been taken to reduce the aging effects on the filter beds:

- (1) Waiting to load the charcoal into the filter as long as possible after painting or using solvents in the building.
- (2) Sparing use of solvents when filters are in place.
- (3) Alternate venting of solvent vapors from building.
- (4) Using "guard beds" replaced at frequent intervals.
- (5) Heating of the beds to drive off the adsorbed highly volatile contaminants.

Table 3. Average ignition temperature for activated carbon without flame retardant material^a

Carbon type	Impregnate	Ignition temperature ^b
Coconut	5% TEDA	190–215°C
Petroleum	5% TEDA	195–240°C
Coconut	2% TEDA	215–330°C
Coconut	None	340°C
Coconut	5% KI	312–333°C ^c

^aBased on Ref. 40.

^bDependent on gas velocity and heating rate.

^cA. G. Evans, Confinement of Airborne Radioactivity Progress Report: January–June 1973, DP-1340 (1973).

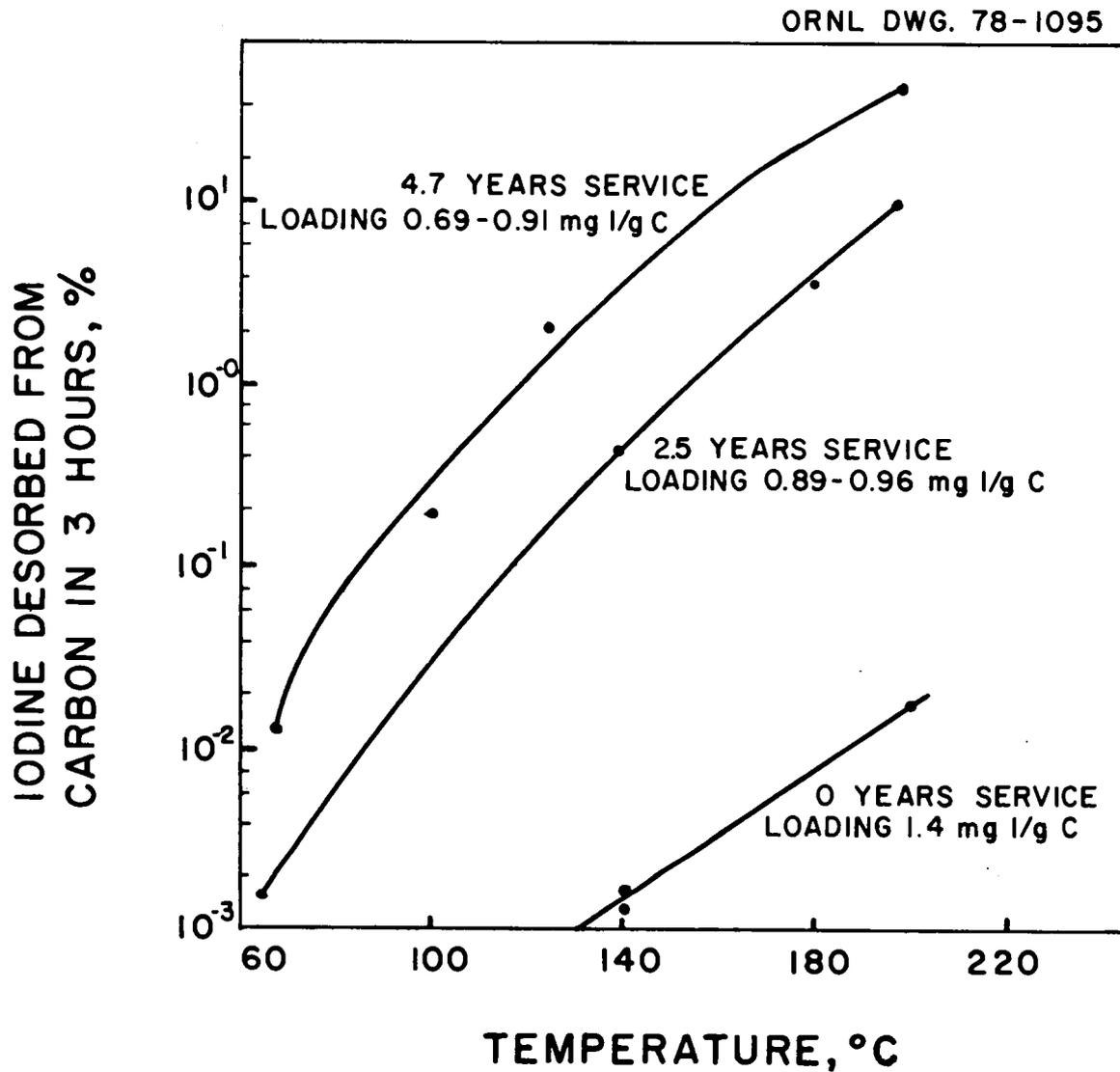


Fig. 4. Increase in desorption with increased temperature and service (from Ref. 41).

5.1.7 Radiation effects

Use in reprocessing plants or in a post-accident environment would subject off-gas or air filters to high dose rates of ionizing radiation. Under these conditions, a portion of the iodine bound to the carbon may be converted to free radicals, which in turn could react with organic contaminants or water and oxidize charcoal itself. These more volatile iodine compounds would readily desorb, lowering the bed's overall efficiency.

An early report by Jones⁴² indicated that an increase in gamma radiation would reduce CH_3I penetration when CH_3I was loaded with nonradioactive elemental iodine onto an unimpregnated carbon bed in a constantly flowing humid air stream. This may have been due to the radiolytic decomposition of CH_3I or enhanced isotopic exchange. It was noted that the presence of carbon significantly inhibits the decomposition of CH_3I , which was assumed to be the effect of reduced mobility of the methyl radicals and iodide ions; thus, the decomposition of CH_3I is a surface reaction and not a gas phase one.

Evans and Jones³⁷ state that since the primary product of radiation induced iodine release is organic iodides, iodine adsorbers impregnated with materials which react with these are more effective than unimpregnated carbon systems.

5.1.8 Relative humidity effects

The operation of a carbon filter in a humid environment (>70% relative humidity) has very detrimental effects on CH_3I removal efficiency and significantly impairs elemental iodine removal if the bed should become saturated. In general, the following statements can be made:

- (1) Water vapor adsorption onto the activated carbon will increase penetration.
- (2) A filter prehumidified with water vapor will have reduced efficiency for CH_3I removal even from dry air.
- (3) Efficiency of a filter to remove CH_3I from moist air will decrease as the adsorbed water in the carbon increases.

- (4) The most adverse removal condition is the use of a prehumidified bed in the presence of a high humidity gas stream.
- (5) As the micropores in the activated carbon are filled with condensed water, the efficiency to remove CH_3I decreases.

Some impregnated carbons show improved iodine removal efficiencies under these conditions if flooding does not occur; flooding of the impregnated carbon caused by condensing steam can wash out major portions of the impregnation. One method to prevent flooding is by preheating the gas stream above its dew point,⁴³ which also results in better removal efficiencies because fewer micropores are filled with condensed water, leaving more active sites.

5.1.9 Concentration effects

The effect of iodine concentration has been investigated by several researchers.^{16,44,45} The early work by Craig,¹⁶ of South Africa, the Idaho Chemical works,⁴⁶ and others pointed to a decrease in removal efficiency as the iodine concentration was lowered.

However, this effect of iodine concentration was also explained by other authors as the result of the existence of a more penetrating form of iodine such as HOI or the generation of such a compound. The following conclusions were drawn from a theoretical analysis by Ritzman and Genco⁴⁷ of the adsorption from a fluid stream flowing through a carbon bed based on five accepted types of isotherms:

- (1) For the ideal case of one adsorbate in an inert carrier gas with no change in species, the efficiency is not a function of concentration in the region where the isotherm is linear.
- (2) For a dynamic experiment, the effect of time could be important.
- (3) Physical effects which cause deviation from the ideal state could be important.

It was also concluded that (1) over the range of 10^{-8} to 10^3 $\mu\text{g}/\text{m}^3$, present experimental or theoretical knowledge provides no firm basis for a strictly concentration-dependent isotherm for either elemental or methyl iodine and (2) the observed variations are the result of one or more secondary effects.

Another problem in removing very low iodine concentrations, shown by Wilhelm,²⁸ is that the natural airborne iodine concentration is on the order of 10^{-3} $\mu\text{g}/\text{m}^3$. Thus, lower iodine concentrations may occur after only passing partially through a filter.

At the 13th AEC Air Cleaning Conference, two papers were published with opposing views on the concentration effects of CH_3I . Kovach and Hunt⁴⁸ reported that removal efficiency for CH_3I in the range of atmospheric concentration did not differ from those for the mg/m^3 range. Bellamy⁴⁹ presented data showing a decrease in removal efficiency for concentrations of $\text{CH}_3^{131}\text{I}$ above 10^{-6} Ci/m^3 on a 1/2-in. deep bed of activated carbon. Deeper beds provide a much less concentration-dependent removal efficiency profile. Unfortunately, there is no indication of the total concentration of iodine used in this study. Wilhelm²⁸ examined the removal efficiencies in the range of activity concentrations greater than 10^{-6} Ci/m^3 , and even at very high activity, he found no concentration effects on bed depths of 2.5 to 10 cm.

5.1.10 Use of oxidizing catalysts

The use of oxidizing beds was proposed first to minimize poisoning of the sorbent bed by catalytic combustion of adsorbable organic vapors and second to improve the filter's efficiency by converting organic iodides to elemental iodine.

Ackley and Adams⁵⁰ showed in 1967 that using chromium salts or iron salts as oxidizing catalysts produced relatively poor performance with respect to removal of CH_3I , which may have been due in part to adsorbed water inhibiting the oxidation process.

With the continuation of research with oxidation beds, Ackley and Davis⁵¹ reported in 1974 that while hopcalite produces slightly improved removal efficiency for CH_3I , it was detrimental to I_2 removal, and the possible benefits of adding a catalytic oxidation bed would not be of sufficient magnitude to warrant the added cost.

5.1.11 Loading capacities

The reported loading capabilities of the carbons vary widely from author to author. Adams et al.⁵² reported loadings in the range of 0.005 to 0.08 mg CH₃I/g of carbon over a wide range of conditions. His work produced removal efficiencies of 99+% for relative humidities less than 70%. Later work by Adams and Ackley⁵³ showed removal efficiencies of 99.998% for a 2-in. bed of either BC-117 or BC-727 when loaded with 4-11 mg I₂/g of carbon at 25°C with a relative humidity of 98%. Data on loadings of 47 to 66 mg I₂/g of carbon showed that a removal efficiency of 99.9+% was achieved for a thin bed at 25°C with a relative humidity of 98%.

Early work by Milham and Jones⁴⁵ indicated that at elevated temperatures (~200°C), after 30 months of service, desorption of iodine from type 416 carbon increased as loading was increased. At a loading of 0.3 mg I₂/g, the carbon desorbed about 8% of the loaded iodine in 1.2 h. Tests at 65°C showed desorption of less than 0.08% for loadings up to 8.9 mg I/g carbon.

A recent report by Wilhelm²⁸ recommended maximum loadings of 100 µg CH₃I/g on KI impregnated carbons and 1 mg CH₃I/g on TEDA impregnated carbons.

Additional information on the behavior of activated carbon can be found in Refs. 54-77.

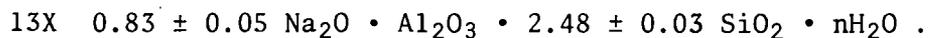
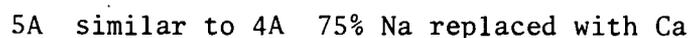
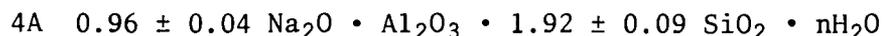
5.2 Zeolites

As mentioned above, carbon filters have certain disadvantages: iodine desorbs at relatively low temperatures, low ignition temperature, possible buildup of explosive gases, and poor performance in a steam environment. A more promising approach to the problem of iodine removal is the use of inorganic adsorbers in which a stable iodine compound is formed. An initial scoping study was performed by Maeck et al.⁷⁸ in which 25 materials were tested for I₂ and CH₃I retention. Based on this study, a synthetic zeolite, Linde molecular sieve 13X, converted to the silver form (AgX) was selected as the most promising of the materials tested.

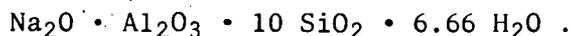
The advantages that AgX has over activated carbon are (1) it is nonflammable, (2) it operates well at elevated temperatures, (3) it poses no explosion hazard, (4) it is resistant to poisoning, and (5) it forms an insoluble iodine product.

5.2.1 Zeolite structure

Different types of zeolites have varying physical properties. One of the most important properties is the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio. The higher this ratio, the harder the material, the more acid resistant the zeolite, and the lower the ion-exchange capability. Hersh⁷⁹ reports the chemical formula of Linde's synthetic zeolites as:



Natural mordenite has the following chemical formula:



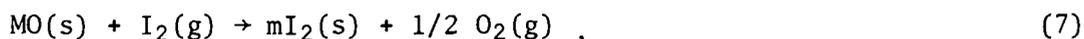
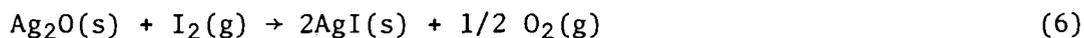
From the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio, the mordenite should be (and is) much harder and acid resistant; however, the X-type zeolite has an ion-exchange capability of about three times that of the mordenite.⁸⁰

The sodium in the zeolite structure can be exchanged for other metal cation. A batch method for silver exchange is presented by Pence and Maeck,^{81,82} and both batch and plug flow techniques are described by Thomas et al.⁸³

5.2.2 Chemical mechanisms

The exact mechanism for iodine sorption on metal-exchanged zeolites has not yet been determined. Physisorption and chemisorption are the two

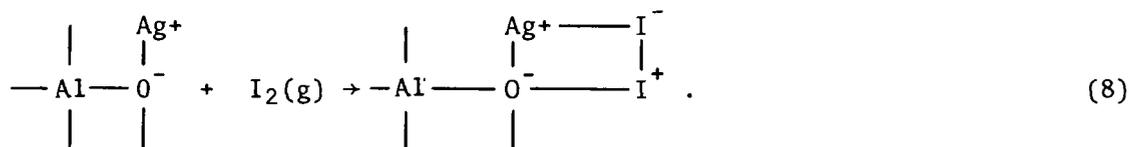
mechanisms that Thomas et al. proposed as responsible for iodine adsorption. The degree to which each of these occurs depends on the type of metal exchange zeolite used (see Table 4 and also Sect. 5.2.3). If one assumes that the metal ions exist as oxides in the zeolite structure, the reaction could be



where

M = Pb or Cd.

The standard free energies of the reactions are -16.1, 0.3, and 2.5 kcal/mole for silver, palladium, and cadmium, respectively. For copper, cobalt, iron, nickel, and zinc, the oxides have free energies of reaction of 20 kcal/mole or more. It is evident that all of the above metal oxides, with the exception of silver, are more stable in their oxide form, which accounts for the large chemisorption capacity of AgX. A plausible mechanism for the reaction might be



Many of the same parameters that affected activated carbon adsorption have been studied on metal-exchanged zeolites. Table 5 lists the important parameters and reports that discuss these parameters.

5.2.3 Use of alternative metal-exchanged zeolites

Due to the high cost of silver, alternate methods using zeolites have been examined to reduce the cost of the metal-exchanged zeolite iodine filter unit. First, other metal cations were exchanged; second,

Table 4. Maximum iodine adsorption capacities
(mg of I₂ per gram of bed)^a
of metal zeolites at 150°C^b

Adsorbent	Saturated	Physisorbed	Chemisorbed
NaX	364	334	30
AgX	349	135	214
PbX	179	153	26
CdX	374	329	45

^aBased on dry weights of 0.61 g/cm³ for NaX,
0.71 g/cm³ for CdX, and 0.85 g/cm³ for PbX and AgX.

^bFrom Ref. 83.

Table 5. Literature reference numbers pertaining to the operation
and behavior of metal-exchanged zeolites

Alternative metals	— 36, 55, 80, 83, 84, 85, 86, 87, 88, 95
Silver Content	— 36, 51, 88
Poisoning	— 36, 59, 80, 81, 82, 83, 88, 96
Regeneration	— 2, 83, 89, 90, 91
Temperature cycling	— 84, 88
Inlet gas concentration	— 55, 80, 88, 91
Relative humidity	— 36, 78, 80, 81, 82, 83, 85, 86, 87, 88, 91
Aging and weathering	— 36, 51, 59, 97
Radiation effects	— 80
NO ₂	— 83, 86, 87
CO ₂	— 88, 96
Temperature	— 61, 83, 85
Steam	— 78, 80, 92
Bed depth	— 55, 80, 83, 97
Particle size	— 51, 75, 80, 82, 88
Gas velocity	— 36, 80, 81, 83, 85

the silver content was reduced; third, a method for filter bed regeneration was developed.

Work by Adams et al. as reported by Unger et al.⁸⁴ on zeolites treated with CrCl_2 , $\text{Pb}(\text{NO}_3)_2$, and $\text{Mn}(\text{NO}_3)_2$ solutions gave CH_3I trapping efficiencies at 200°C of 6, 32, and 68%, respectively, for loadings of $0.5 \text{ mg CH}_3\text{I}/\text{cm}^3$ sorbent on a 2-in. deep bed. Thus, none of these materials warranted additional work. Studies were also performed on cadmium and potassium zeolite at 200°C at higher loadings. These efficiencies decreased rapidly as loadings were increased to $37 \text{ mg}/\text{cm}^3$; the limit of chemisorption had been reached at this loading. The idea of a mixed bed of AgX and another metal-exchanged zeolite to extend the useful life of the more costly silver zeolite was also proposed.

Maeck and Pence⁸⁵ studied lead-exchanged zeolite and found that while PbX had a low removal efficiency for CH_3I , more than 97% of elemental iodine was removed from air at 21% and 90% relative humidity at bed temperatures above 45°C .

Later work by Pence et al.⁸⁰ examined additional metal cation-exchanged zeolites: cadmium, copper, mercury, palladium, thallium, and some rare earth cations. Some showed promise for elemental iodine removal, but efficiency decreased markedly with increasing relative humidity at a fixed temperature. In spite of the increase in water content in the air, the adsorption efficiencies also increased as the bed temperature was increased from 22 to 90°C at fixed relative humidity. Mercuric ion-exchanged Y-type zeolite, Hg^{2+}Y , showed the best efficiency of all zeolites tested other than silver. It was capable of removing more than 99.9% of the I_2 from a water saturated air stream at 22°C . For CH_3I , the efficiency of the metal-exchanged zeolites, except silver, decreased significantly. The highest was Hg^{2+}Y with a removal efficiency of 33.8% at 125°C and 90% relative humidity. PbX also showed some promise in a heated bed for CH_3I removal.

Mailen, as reported by W. D. Burch et al.,^{86,87} examined the effects of NO_x on potassium-, cadmium-, and palladium-exchanged zeolite beds. Tests showed less than 2.5% removal efficiency of CH_3I for potassium and cadmium zeolites and less than 0.25% removal of I_2 for palladium zeolite

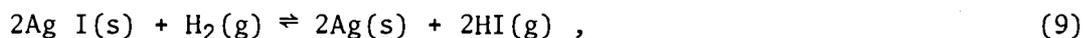
in the presence of 2% NO_x. These tests also showed a reduction in efficiency with water vapor present.

Thomas et al.⁸³ also examined metal substituted zeolites. Cadmium and palladium were effective up to loadings of 3.5 mg I₂/g of substrate, but copper and zinc zeolites were found ineffective. Studies with the sodium form of the zeolite indicated that it was the substrate that traps the iodine and not the palladium or cadmium. Silver-exchanged zeolites, however, gave removal efficiencies of 99.5% at loadings up to 190 mg I₂/g. As suggested above, only metals that have iodine forms more stable than the oxide form would be potential substitutes for silver.

Somewhat more limited work has been done on partial exchange of silver for sodium to reduce the filter cost. Pence, Duce, and Maeck³⁶ reported that the adsorption efficiency of AgX in a high humidity environment was quite sensitive to the extent of silver exchange. Ackley and Combs⁸⁸ reported on zeolites with 10 to 51% of the sodium exchanged for silver. Twenty-seven percent AgX appeared to have acceptable performance at 200°C. With a loading of 13.1 mg I₂/cm³ 27% AgX yielded a DF of 4.6 × 10³. Ackley and Davis⁵¹ indicated that the size of the particle was more influential than the amount of silver exchange, for example, 26% AgX 1/16-in. pellets were superior in performance to 88% AgX 1/8-in. pellets.

5.2.4 Regeneration of AgX

After limited success in attempts to develop an alternate metal-exchanged zeolite to replace the costly silver form, studies were undertaken to find a method to regenerate the filter bed. Work at Idaho National Engineering Laboratory^{83,89} showed that regeneration using high temperature (400-500°C) hydrogen was possible. A flow plan of the process is shown in Fig. 5. The proposed reaction is:



ORNL DWG. 78-1594

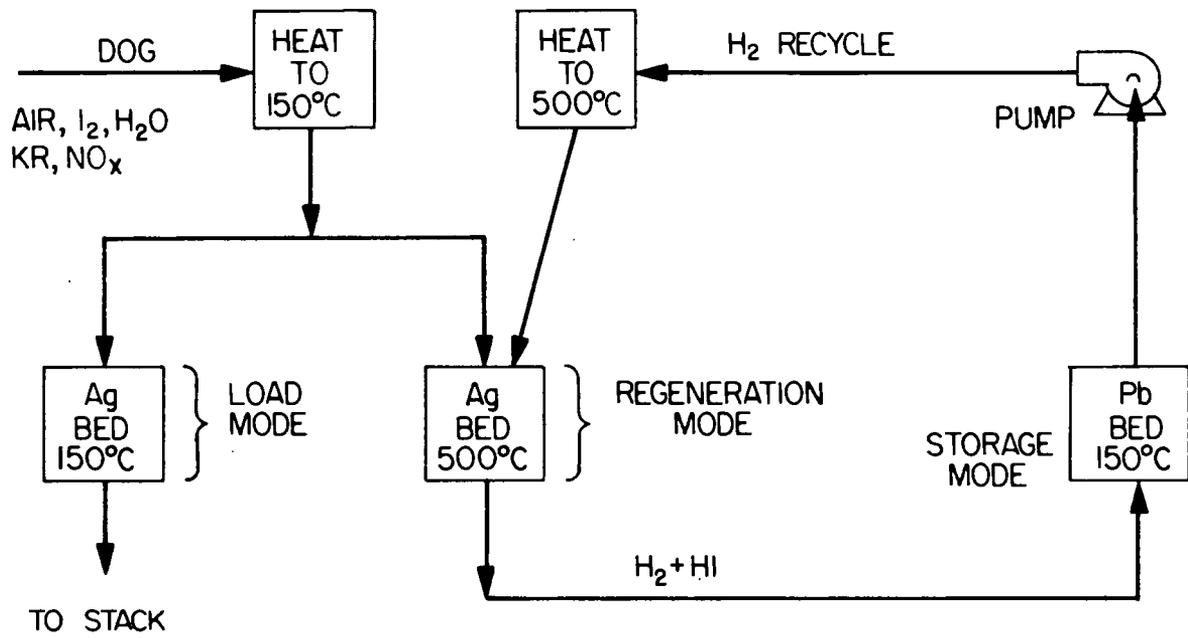


Fig. 5. Block diagram of iodine recovery process (from Ref. 83).

with the HI produced being chemisorbed on the lead zeolite. As mentioned above, the chemisorption of I_2 in PbX is not thermodynamically favored; however, the reaction



has a free energy of reaction of -47 kcal/mole at 150°C. It was reported that at a desorption temperature of 500°C, the beds can be regenerated ten times faster than the projected iodine adsorption rate. Thermodynamic data for HI desorption has been generated by J. C. Mailen⁹⁰ and may be found in Appendix C for various temperatures.

Regeneration temperatures of 400 to 600°C have been tested. Little effect is noted between 400 and 500°C, however, there was a two-fold loss in capacity of the AgX after five cycles at both temperatures. After three cycles at 600°C, there was a threefold decrease in capacity, which was attributed to pore collapse within the zeolite structure; therefore, a limit of 500°C is recommended for zeolite regeneration.

The iodine removed from the AgX bed was chemisorbed onto lead as PbI_2 loadings to 88% utilization of the lead (~ 389 mg I_2 /g PbX) have been obtained. A bright yellow compound, PbI_2 , has a solubility in water of 26 mg I_2 per 100 ml of water at room temperature.

Work by Burger et al.² was directed toward the fixation of HI in bismuth and copper solutions. Copper solutions required the introduction of SO_2 to prevent the carry through of I_2 from the reactions



Bismuth solutions were then employed to avoid the reoxidation problems of the copper solution. The final product BiI_3 was a fine, dense, black precipitate. A problem associated with this process was that BiI_3 , in some cases, held excess I_2 which was volatilized upon heating.

Early work by the association S.C.K./C.E.N-Belgonucleaire⁹¹ indicated that nonexchanged zeolites could be regenerated five times at 200°C with no effect on capacity. However, as pointed out above, the major portion of the iodine is physically adsorbed rather than chemically adsorbed; no mention was made of the exact method of regeneration or the medium for final iodine trapping.

5.2.5 Poisoning of AgX

The effects of various contaminants have been studied by several workers. Ackley and Combs⁸⁸ examined the influence of dodecane and found no influence at 100°C, but as the temperature was increased from 100 to 350°C, the efficiency decreased. This decrease was attributed to the formation of tars on the zeolite. Improved efficiency was noted at 450°C, possibly indicating decomposition of the tars. Studies on CO₂ showed no effects on efficiency. Chlorine caused deleterious effects which are expected for other halogens and halogen containing compounds. This effect was noted in the *Nuclear Air Cleaning Handbook*⁵⁹ where HCl vapor seriously degrades the zeolite trapping efficiency.

Pence et al.⁸⁰ examined the effects of several gases on the methyl iodide removal efficiency of silver zeolites (see Table 6). Of the gases studied, only H₂S and SO₂ had serious effects. Small adverse effects were noted for both NO₂ and propane, however, these studies were carried out under abnormally high contaminant gas concentrations.

Work by Thomas et al.⁸³ indicated a three-fold decrease in I₂ loading on AgX in the presence of 2% NO₂. Also noted was the interaction of water vapor with the NO₂.

5.2.6 Temperature cycling

Temperature cycling of silver zeolite exposed to humid air, a potential damaging process, was studied at ORNL.^{84,88} The study involved cycling the sorbent between 200 and 28°C and then measuring the efficiency of the sorbents at 200°C. Tests indicate that a moderate number (2 or 3) of cycles do not seriously affect the removal efficiency.

Table 6. Effects of contaminant gases on the methyl iodide adsorption efficiency of silver zeolite^a

Conditions: 90°C, 90% relative humidity, 23 cm/sec face velocity, and 0.1 µg/m³ of methyl iodide

Contaminant gas	Contaminant concentration (vol %)	Adsorption Efficiency (%)
O ₂	10	99.94
CO ₂	10	99.95
NO ₂	10	99.79
H ₂	1.9	99.97
H ₂ S	0.06	42.20
SO ₂	1.9	31.48
Propane	10	99.89
Hexone	2.2	99.99
Kerosene	0.05	99.91

^aFrom Ref. 80.

5.2.7 Concentration effects

Pence et al.^{55,80} reported an apparent concentration effect on AgX which was more noticeable at lower gas velocities where the removal efficiency was higher. The effect may be the result of a change in form of iodine into a more penetrating type as was the case for carbon. This occurred at concentrations in the range 10^{-5} to 10^{-4} $\mu\text{g CH}_3\text{I}/\text{m}^3$.

Ackley and Combs⁸⁸ indicated an apparent concentration effect on low silver-exchanged zeolites. This effect became less distinct at high degrees of exchange. A rather limited range of concentrations, 21 to 160 $\text{mg CH}_3\text{I}/\text{m}^3$, was examined in this study. The association S.C.K./C.E.N.-Belgonucleaire report⁹¹ showed no concentration effect for I_2 concentrations of 70 to 400 $\text{mg I}_2/\text{m}^3$ on 13X molecular sieves.

5.2.8 Temperature effects

An increase in temperature increased the removal efficiency of AgX at all relative humidities. One explanation presented by Lorenz et al.⁶¹ was that the increase in temperature improves the gas-phase mass transfer.

While extended exposure to a steam environment can cause a physical breakdown of the zeolite structure,⁹² zeolites are far more effective (99.5% removal efficiency)⁸⁰ than carbons in the steam environment.⁷⁸ Steam exposure did, however, cause a slight reduction in efficiency (99.9 to 99.5%).

5.2.9 Relative humidity

The effects of relative humidity have been studied by numerous researchers, and consistent results have been obtained. While there was a decrease in efficiency with increased humidity, the adverse effects were far less than those for carbon. The decrease in efficiency was also less at higher temperatures. For a fixed relative humidity, CH_3I adsorption increased with temperature.⁸⁵

5.2.10 Miscellaneous effects

The effects of other parameters can be summarized as follows:

- (1) An increase in bed depth increased the iodine removal efficiency.
- (2) The smaller the filter particle size, the better the removal efficiency and loading per unit weight.
- (3) As the gas velocity through the bed increased, the efficiency decreased.

Additional information on the behavior of zeolites can be found in Refs. 93-97.

5.2.11 Process capacity

Obtaining the maximum operational process capacity of silver zeolite is an almost impossible task because of the number of variables concerned. The maximum theoretical capacity is 1192 mg I₂/g of silver,⁸⁹ and at 100% exchange of silver for sodium the silver would compose 36%⁸³ of the structure; therefore, the maximum loading per gram of silver zeolite would be 429 mg I₂. There have been many test runs at low loadings (<10 mg I₂/g AgX), and decontamination factors have been obtained ranging from 10² to 10⁵. A summary of high loading tests is presented in Table 7.

Pence et al.⁸² showed that a DF of 10⁴ was possible for I₂ removal at 22°C on a 5.08-cm deep bed with a gas velocity of 92 ft/min under 100% relative humidity conditions. The gas concentration was 1 to 10 µg I₂/m³. At high gas concentrations (~100 mg I₂/m³), Ackley and Combs⁸⁸ achieved DF's greater than 10⁵ for AgX at 200°C with gas velocities of 63 ft/min and low relative humidity (~0.2%). These were for relatively low silver utilizations (1.52-4.7%). This report showed that there are only slight differences in removal efficiencies for elemental iodine in the range of 100 to 200°C; however, at 26°C with the same water content in the air (3.0 vol. %, i.e., 90% relative humidity at 26°C), the DF for CH₃I at low loadings (~10 mg CH₃I/g AgX) was only 18, and the gas velocity was 40 ft/min. In dry air with the same conditions, the DF for CH₃I was greater than 10⁴.

Table 7. Summary of high loading capacities of AgX and AC-6120

Reference	Material	Iodine species	H ₂ O (%)	NO ₂ (%)	Temperature (°C)	Capacity ^a	DF
83	AgX	I ₂	0	0	100	139	NR ^b
83	AgX	I ₂	5	2	200	55	NR
83	AgX	I ₂	NR	NR	150	214	NR
82	AgX	CH ₃ I	0	0	25	84	>10 ⁴
88	AgX	CH ₃ I	3	0	200	209	>100
88	AgX	CH ₃ I	3	0	200	87	>10 ⁴
83	AgX	I ₂	NR	NR	30-100	52	NR
93	AC-6120	CH ₃ I	3	2.5	150	40	>100
94	AC-6120	I ₂	NR	NR	NR	85	>10 ⁴

^a mg of species/g of substrate.

^b Not recorded.

5.2.12 Advantages and disadvantages

Silver zeolite can be used in a wide range of operating conditions with excellent results for CH_3I and I_2 removal. Its advantages can be summarized as:

- (1) Low flammability.
- (2) Low explosion hazard.
- (3) Final silver zeolite-iodine complex is a solid for final storage.
- (4) Removal efficiency increases with temperature and shows efficient sorption to 500°C .
- (5) High temperature (up to 1000°C) retention.
- (6) Removal of elemental iodine not affected by many contaminants.
- (7) High capacity for CH_3I .

Silver zeolite also has the following disadvantages:

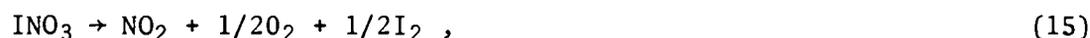
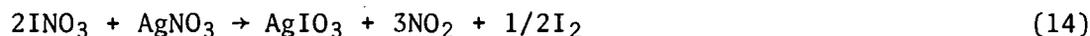
- (1) The cost of silver makes the product about 20 times as expensive as carbon.
- (2) X-type zeolites are not acid resistant and are also affected by prolonged exposure to steam.
- (3) CH_3I removal is affected by SO_2 , H_2S , and to some degree C_3H_8 and NO_2 .

5.3 Alumina Silicates

Another class of inorganic sorbents for iodine and methyl iodide removal is silicic acid and aluminum oxide to which silver nitrate has been added. The West Germany iodine removal effort has been developed around a once-through process using AgNO_3 impregnated amorphous silicic acid (under the trade name AC-6120), which is commercially available in Europe. Developmental work in the United States has involved North American Carbon Company's GX-135, a silver nitrate impregnated alumina silicate. Work has also been subcontracted to the University of Belgrade, Yugoslavia by the U. S. Environmental Protection Agency to examine the potential for iodine removal for various nitrates on Alcoa alumina H-151.

5.3.1 Chemical mechanism

The silver nitrate in the alumina silicate structure reacts with elemental iodine by²⁸



where the silver binds iodine as either silver iodide or silver iodate. Alkyl halides also react with the silver to form silver iodide



The end products of both reactions with silver are stable solids with low solubilities in water.

5.3.2 H-151

A limited amount of information is available on the Yugoslavian work with H-151.⁹⁸ The chemical composition of H-151 is given in Table 8. Tests have been performed with impregnations of the following metal nitrates: Ag, Cu, Pb, Cd, Zn, Ni, Co, Ce, Ag + Cu, Ag + Ce, Ag + Cd. The metal salt contents are shown in Table 9. Of these metal nitrates, only silver, and to lesser degree copper and lead, showed irreversible chemisorption of methyl iodide at 56°C. The capacity of cadmium nitrate to bind CH₃I increased at 96°C to nearly that of silver nitrate in dry air.

Thermodynamic calculations indicated that silver oxide, AgO, might be a better choice than silver nitrate and that silver nitrate is better than the other metal oxides tested. Later studies revealed that AgO was superior only in a dry environment.

Table 8. Specification of base alumina^a

Base adsorbent: Alcoa alumina H-151

Composition, %	
Al ₂ O ₃	90.0
Na ₂ O	1.4
Fe ₂ O ₃	0.1
SiO ₂	1.7
Loss on ignition (1100°C)	6.0
Surface area, m ² /g	390
Bulk density, packed, lb/ft ³	53
Specific gravity	3.2
Particle sizes, cm	0.34 and 0.12

^aFrom Ref. 98.

Table 9. Impregnations of alumina^a

Metal salt content of impregnated H-151 alumina samples

No.	Salt(s) impregnated	Millimoles per 100 g	Metal (wt %)
1	AgNO ₃	78.5	8.47
2	AgNO ₃	42.0	4.53
3	Cu(NO ₃) ₂	80.3	5.10
4	Cu(NO ₃) ₂	41.0	2.60
5	Pb(NO ₃) ₂	60.0	12.43
6	Pb(NO ₃) ₂	39.0	8.08
7	Ni(NO ₃) ₂	78.1	4.57
8	Ni(NO ₃) ₂	42.0	2.46
9	Co(NO ₃) ₂	80.0	4.72
10	Co(NO ₃) ₂	42.0	2.48
11	Cd(NO ₃) ₂	68.0	7.64
12	Cd(NO ₃) ₂	43.0	4.83
13	Zn(NO ₃) ₂	76.0	4.96
14	Zn(NO ₃) ₂	50.0	3.26
15	Ce(NO ₃) ₃	50.0	7.00
16	AgNO ₃ + Cu(NO ₃) ₂	41.0 + 41.0	4.42 + 2.60
17	AgNO ₃ + Ce(NO ₃) ₃	38.0 + 24.0	4.09 + 3.36
18	AgNO ₃ + Cd(NO ₃) ₂	10.0 + 17.1	1.08 + 1.92

^aFrom Ref. 98.

Tests also showed that the presence of water vapor decreased the CH_3I adsorption efficiency, which has been seen in other systems. An increase in velocity also decreased removal efficiency.

The best results obtained for AgNO_3 impregnated alumina (7.4 wt % Ag) with a particle size of 0.08 to 0.16 cm was 99.9% retention with a face velocity of 8 cm/sec and a relative humidity of 27% at 50°C. The CH_3I loading was 1.9 mg/g of adsorbent.

5.3.3 GX-135

Work on North American Carbon Company's silver-containing alumina oxides has been carried out at ORNL by Ackley and Combs⁸⁸ and Ackley and Davis.⁵¹ These products include GX-100, a ball-shaped silver containing alumina of either 8 × 16 or 16 × 30 mesh; GX-135, a granular, 8 × 16 mesh, silver salt treated combination of aluminum and silicon oxides; and GX-148, similar to GX-135 except that it is 16 × 30 mesh.

A removal efficiency of 99.997% was obtained for 16 × 30 mesh GX-100 at 100 and 200°C for low loadings, 0.4 mg $\text{CH}_3\text{I}/\text{cm}^3$ sorbent. However, at higher loadings (13-46 mg $\text{CH}_3\text{I}/\text{cm}^3$) the removal efficiency dropped from 58 to 76.5%. It was moderate efficiency for removing butyl iodine if the loadings remained low. GX-100 was very inefficient (12.9% removal efficiencies) at 27°C with 3% water vapor.

GX-135 was highly effective (>99.9997) in removing elemental iodine at 100 and 200°C if the iodine loadings remained at 5 mg/ I_2 or less. GX-135 offered high efficiencies greater than 99.96% for loadings up to 10.9 mg $\text{CH}_3\text{I}_2/\text{cm}^3$ with 3% water at 200°C. A high efficiency greater than 99.995 was also obtained for lower loadings less than 6.3 mg $\text{CH}_3\text{I}/\text{cm}^3$ at 100°C with 3% water.

At 100 and 255°C, high efficiencies for GX were achieved for 100 and 200°C with loadings up to 11.4 mg $\text{CH}_3\text{I}/\text{g}$. GX-148 performed somewhat better than GX-100 at 27°C and 3% water, yielding a 56.1% removal efficiency; however, 90% AgX showed a removal efficiency of 96.3% under the same conditions, i.e., sorbent loading of 12.0% mg $\text{CH}_3\text{I}/\text{cm}^3$ and 3% water. Tests on GX-148 were discontinued because of handling difficulties with the fine mesh size.

5.3.4 AC-6120

In 1970 J. G. Wilhelm and H. Schuttelkopf⁹² introduced the use of several catalyst carriers impregnated with AgNO_3 as materials to remove iodine from gas streams at elevated temperatures and from gas streams containing acid vapor. These carriers (KTB, KTC, KTD) were experimental products from Bayer and had a porosity greater than 50% and a specific surface of about $110 \text{ m}^2/\text{g}$.

Early experiments using a bed depth of 10 cm showed high removal efficiency (>99.9944%) for CH_3I on 1.6-mm pellets with an impregnation of $\leq 7.8 \text{ g Ag}/100 \text{ g sorbent}$ and CH_3I loadings of 1 to 2 mg $\text{CH}_3\text{I}/\text{g sorbent}$ at 30°C and a relative humidity of 70%; however, at 100% relative humidity, the removal efficiency dropped to 27%. Tests using superheated steam showed removal efficiencies greater than 99.9984%. High removal efficiencies (>99.44) were also obtained for KTC impregnated with 7.8 to 10.4 g $\text{AgNO}_3/100 \text{ g}$ from a gas stream containing 8 to 10% NO_2 . The removal efficiency varied directly with the bed depth and inversely with relative humidity and loading. It was also noted by the workers that an iodine compound more penetrating than CH_3I was also present and that KTC material had a very low efficiency for its removal. Initial economic studies indicated that due to the low silver content of KT materials (18 or less g $\text{Ag}/100 \text{ g sorbent}$), the cost would be about three times more than impregnated carbon and nearly ten times less than silver zeolite.

In later studies,⁹³ Ag-KTB was shown to have higher mechanical strength and abrasion resistance than the other KT products. Ag-KTB has since become available under the product number AC-6120. Both Ag-KTB and Ag-KTC have similar removal efficiencies with equal AgNO_3 impregnation. The normal impregnation was 60 to 80 mg $\text{Ag}/\text{g KT material}$. It was recommended due to the detrimental effect of water vapor that 70% relative humidity should be the practical limit in operation. It was also shown that 150°C was the optimum operating temperature. There is, however, a slight increase in removal efficiency above 200°C , but operation above did not appear cost effective. Additional tests were run with NO_2 . The results indicated a small decrease in efficiency from 99.99961% to 99.9973% for 1% NO_2 to 10% NO_2 , respectively. Loadings in excess of

10 mg $\text{CH}_3\text{I}/\text{g}$ AC-6120 resulted in drastic decreases in removal efficiency. The removal efficiency of AC-6120 did not seem to be influenced by irradiation up to 8.6×10^8 rd.

In a paper presented at the 1976 ERDA Air Cleaning Conference, Wilhelm⁹⁴ discussed the application of AC-6120 in the role of iodine removal for a fuel reprocessing plant. Also included were test results from the high impregnation version AC-6120/ H_1 with 12 wt % silver as compared to AC-6120 with 7 wt % silver. If 100% of the silver reacts, 143g ^{129}I could be removed by 1 kg of AC-6120/ H_1 , however, 75 to 94% utilization is normal. Decontamination factors of 10^4 were usually achieved for loadings of about 60% utilization.

Tests were run to examine the effects of various organic iodides (see Table 10). Primary, secondary, and tertiary alkyl iodides all reacted, but the rate of reaction decreased with chain length, and tertiary alkyl iodides reacted much faster than primary or secondary alkyl iodides. Iodocyclohexane and benzyl iodide both reacted at temperatures up to 180°C with the silver NO_3 in AC-6120.

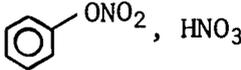
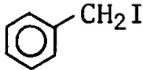
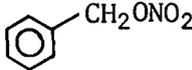
Studies using dodecane and tributyl phosphate (TBP) which are expected in the vessel off-gas indicated that dodecane was completely inert with respect to AC-6120, and TBP was converted to a silver phosphate of unclarified structure. AC-6120 also showed signs of extensive discoloration accompanied by a substantial decrease in efficiency which was attributed to conversion of the AgNO_3 to Ag_2O . A NO_2 air mixture eliminated the discoloration and largely restored the efficiency. A low NO_2 content in the off-gas was also able to control the poisoning of AC-6120 filters. The silver in AgNO_3 was reduced to metallic silver by NO , and the removal efficiency was also reduced by NO . This effect was also reduced by the presence of NO_2 .

Other studies in West Germany⁹⁹ indicated that in long-term tests, AC-6120 was useful only in temperatures below 250°C .

5.3.5 Process capacity

For AC-6120/ H_1 , the maximum theoretical loading is 143 mg I_2/g AC-6120/ H_1 ; for AC-6120, the maximum loading is 73 mg I_2/g AC-6120. The

Table 10. Reactions of AC-6120 with gaseous organic iodine compounds^a

Compound	Reaction products
CH ₃ I	AgI, CH ₃ OHO ₂ , alkanes
C ₂ H ₅ I	AgI, C ₂ H ₅ ONO ₂
C ₁₂ H ₂₅ I	AgI, C ₁₂ H ₂₅ ONO ₂
CH ₃ -CHI-CH ₂ -CH ₃	AgI, H ₃ C-CH=CH-CH ₃ ,
(CH ₃) ₃ CI	AgI (CH ₃) ₂ -C=CH ₂ , C _n H _{2n+1} I
	AgI,  , HNO ₃
	AgI, 
	No reaction up to 180°C

^aFrom Ref. 94.

7.4% AgNO_3 impregnated H-151 has a theoretical capacity of 88.2 mg I_2/g sorbent. Actual iodine loading of AC-6120/ H_1 and AC-6120 should be limited to 10 mg $\text{CH}_3\text{I}/\text{g}$ sorbent. To achieve the high removal efficiency, G-135 should be limited to 5 mg I_2/g sorbent. H-151 required an even lower load limit of 2 mg CH_3I to achieve removal efficiency above 99.9%.

5.3.6 Advantages and disadvantages

Alumina silicates offer many of the same operating advantages of silver zeolites and some unique to the material. These are summarized as follows.

- (1) Iodine is chemisorbed to the silver in a solid suitable for long-term disposal.
- (2) AC-6120 and AC-6120/ H_1 can achieve high silver utilization, thus, making the sorbent cost attractive.
- (3) AC-6120 and AC-6120/ H_1 are not adversely affected by NO_2 .
- (4) The NO_2 from the dissolver will serve to partially protect the AC-6120 or AC-6120/ H_1 filter from poisoning.
- (5) High removal efficiency of AC-6120 and AC-6120/ H_1 results when in a superheated steam environment.
- (6) H-151 offers good removal efficiency at low temperatures.
- (7) AC-6120, AC-6120/ H_1 , and GX-135 offer high removal efficiencies between 100 and 200°C; 150°C is the optimum for the AC-6120 and AC-6120/ H_1 filter.
- (8) Radiation does not adversely affect the efficiency of AC-6120.

The disadvantages are:

- (1) H-151 is adversely affected by relative humidity at lower temperatures.
- (2) AC-6120 and AC-6120/ H_1 are adversely affected by some organics.
- (3) AC-6120 shows poor removal efficiency above 70% relative humidity.
- (4) AC-6120 should not be used above 250°C for extended periods of time.²⁹

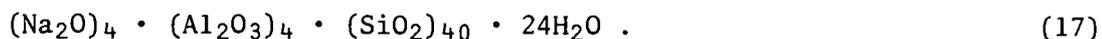
5.4 Mordenites

The use of silver-exchanged mordenites began as an extension of the metal-exchanged zeolite developmental work. Silver-exchanged adsorbents appeared well-suited for bulk iodine removal except that their high cost would make one time use almost prohibitive on a large scale. As a result, other less expensive metal substituted zeolites were examined to determine their usefulness in iodine removal. These zeolites proved ineffective, and developmental work on a method of regeneration for silver sorbents was begun. One of the primary objectives of this program was to identify a substrate with high thermal stability and a substrate that would retain its iodine removal capacity after multiple cycles.

As mentioned above, silver zeolite lost about 50% of its iodine capacity after five regenerations and was also adversely affected by acid vapors. Literature from Norton Company indicated that zeolon molecular sieve material has superior acid-resistant characteristics compared to X- and Y-type zeolites¹⁰⁰ (see Fig. 6).

Zeolons are a family of crystalline aluminosilicate compounds, both natural and synthetic, with extremely uniform pore openings and a high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio; the high $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of ten is indicative of high chemical stability. The precise structure of mordenites contrasts strongly with the wide pore size spectrum found in conventional amorphous adsorbents such as silica gel, activated carbon, and activated alumina.

Zeolon 900 is a self-bonded synthetic mordenite. The lack of an added bonding agent is beneficial to its chemical stability. Zeolon 900 has the following chemical formula in the sodium form:



The effective pore diameter is 8 to 9 Å, and the material has a specific surface of 400 to 450 m²/g.

5.4.1 Chemical mechanism

The chemical reactions that occur on the mordenite with elemental iodine and organic iodides have not been fully explained; however, it

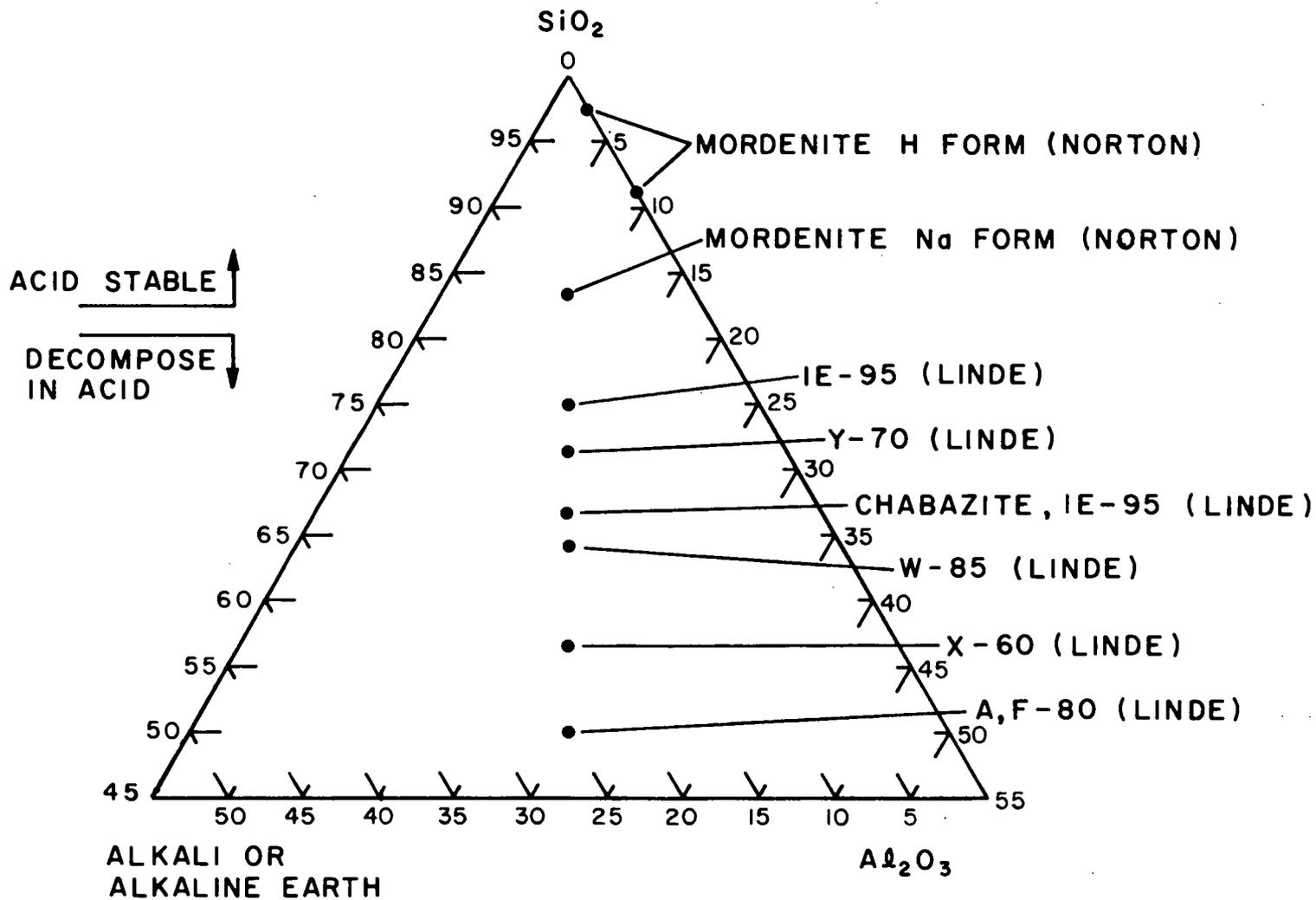
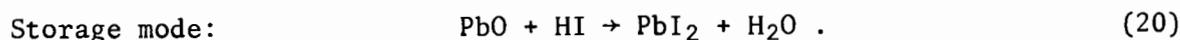
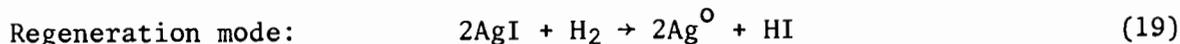
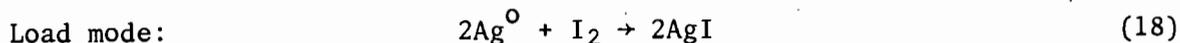


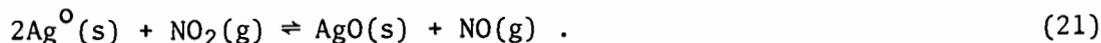
Fig. 6. Composition diagram of molecular sieves indicating acid stability. Axes are in mole % (from Ref. 100).

has been noted⁸³ that the silver exists in both the ionic and metallic states. It also appears that the metallic state has a higher capacity than the ionic state for iodine chemisorption.

A proposed set of reactions for silver and iodine on mordenite is



It was reported¹⁰¹ that NO had a positive influence on iodine loading. In the absence of NO, NO₂ had a negative influence which may be due to the equilibrium reaction

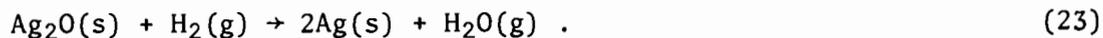


Neglecting the effects of the mordenite matrix, the free energy of reaction to convert the metal to oxide is 6.3 kcal/mole at 150°C. This is thermodynamically unfavorable and would only occur in the absence of NO; NO also acts to hold silver in the metallic state even in the presence of O₂.

Extended purges of 16 to 64 h with dry air at 100°C resulted in a 60% decrease in the capacity of the silver mordenite.⁸³ The equilibrium reaction



may be involved. At 180°C the free energy of reaction to the oxide is zero. The addition of H₂ could protonate the oxygen atom via the irreversible reaction



5.4.2 Engineering data and experimental results

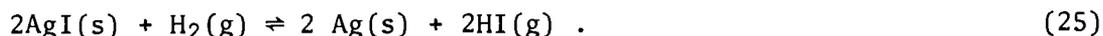
The initial studies on silver mordenite at Idaho were reported by Slansky et al.⁸⁹ in 1976. This report indicated a 50% reduction in capacity of silver zeolite on five regenerations at 500°C with pure hydrogen while at the same time no reduction in iodine capacity was noted for silver-exchanged mordenite-zeolon 900(AgZ). The regeneration method was the same as described earlier for AgX.

In the progress report for the next quarter, Murphy et al., as reported by Slansky et al.,¹⁰² reported loadings of the AgZ of 59.5 ± 6.7 mg I₂/g AgZ (at 95% confidence level) with a DF of 250.

Desorption studies indicated that partial pressure of HI could be approximated by

$$\log P_{HI} = -\Delta G^\circ / 4.6 RT \quad (24)$$

for the reaction



The rate of desorption of HI from AgIZ was modeled by the second order equation

$$Y = 0.585 + 0.456 X + 0.477 X_2 + 0.064 X_1^2 + 0.0045 X_2^2 + 0.352 X_1 X_2, \quad (26)$$

where

$$Y = \text{mg HI/min-cm}^2,$$

$$X_1 = (\text{temp} - 475)/75$$

$$X_2 = (\text{face velocity} - 369)/246.$$

The observed desorption rates agreed within $\pm 11.1\%$ (at 95% confidence level) of the rates predicted.

A later report by Nichols et al., as reported by Slanskey et al.,¹⁰³ indicated the deleterious effect of a 16 to 64 h dry air purge at 100°C. This 60% decrease in loading capacity could be eliminated by the pretreatment of silver with H₂. Results also indicated that effects of the dry air purge could be removed by operating at 150°C.

Additional testing of the regeneration of the AgZ was also reported by Nichols in a report by Slanskey et al.⁹⁵ Typical loading and regeneration operating conditions are shown in Table 11. The beds were loaded with about 35 g I₂, and then more than 99% of the iodine was stripped in each regeneration. Decontamination factors of 10⁴ to 10⁵ were achieved with loadings of 187 mg I₂/g AgZ in the saturation zone of the bed.

Tests of the PbX beds indicated that loadings of 400 mg I/g PbX were possible. The maximum loading for PbX is 440 mg I/g PbX; thus, 90% of lead was being used.

The final report on AgZ development from Idaho National Engineering Laboratory¹⁰¹ stated that the AgZ bed had been loaded with I₂ and recycled 13 times with a 20% loss in capacity compared to the initial loading (see Fig. 7). The increased capacity on the second recycle was attributed to additional silver being exposed and reduced during the first regeneration. The progressive decrease in capacity was explained as a slow but steady pore collapse and decreased accessibility to the silver atoms.

It was also reported that silver oxide is not formed above 100°C in Ag⁰Z except by the reaction with NO₂ (Eq. 21), thus, no effect due to higher bed temperatures would be observed. Also noted was that other gaseous elemental halides should reduce the capacity of Ag⁰Z, however, no laboratory test results were presented to verify this statement.

5.4.3 Process capacity

The maximum theoretical loading of AgZ is 237 mg I₂/g AgZ. Regularly achieved in the saturation zone of the filter beds was 187 mg I₂/g AgZ, which represents about 79% conversion of the silver. The average loading for the bed was obtained as 100 to 130 mg I₂/g AgZ. The maximum loads achieved on PbX have been approximately 400 mg I₂/g PbX which was about

Table 11. Conditions for recycle tests on Ag^oZ^a

Experimental variable	Iodine loading	Iodine stripping
Bed diameter (cm)	5	5
Bed depth (cm)	15	15
Particle size (mesh)	10-20	10-20
Superficial face velocity (m/min)	15	15
Bed temperature (°C)	150	500
Inlet pressure (mm Hg°)	700	760
Carrier gas	air	hydrogen
Iodine concentration at 21°C and 1 atm (mg/m ³)	1500 ^b	7400
NO ₂ concentration (%)	2	0
NO concentration (%)	2	0
Dew point (°C)	35	nil
Iodine flux to and from bed (mg min ⁻¹ cm ⁻²)	1.5 ^c	4.5

^aFrom Ref. 101.

^bActual dissolver off-gas (DOG) concentration is anticipated to be about 380 mg of I₂/m³.

^cActual DOG iodine flux would be about 0.4 mg min⁻¹ cm⁻².

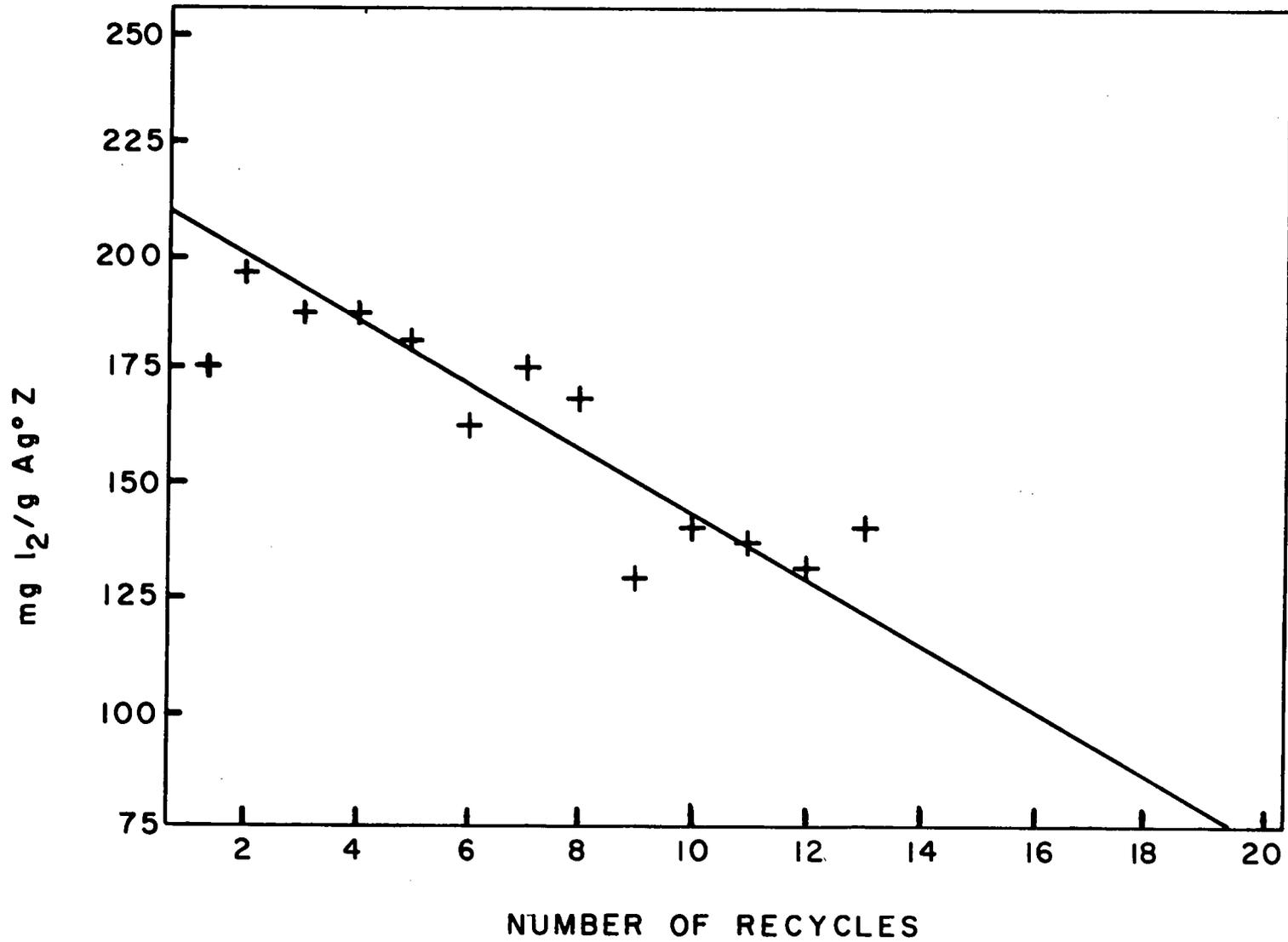


Fig. 7. Iodine loadings of recycled Ag⁰Z in the saturation zone (from Ref. 102).

90% of the theoretical loading. Lead-exchanged mordenite (PbZ) was also tested as a final bed. PbZ has a maximum theoretical loading of 247 mg HI/g PbZ as PbI_2 ; however, loadings as high as 398 mg HI/g PbZ have been reported,¹⁰¹ which may have been the result of the formation of H_2PbI_4 .

5.4.4 Advantages and disadvantages

Mordenites have many of the same characteristics and advantages of zeolites. In addition to those, mordenites also have the following characteristics:

- (1) The capacity for I_2 is improved by the presence of NO in the gas stream.
- (2) A high recycle capability; 13 recycles with only a 20% reduction in loading capacity.
- (3) More acid resistant than zeolites.
- (4) The adverse effects of 2% NO_2 and/or 6% H_2O is negated by 2% NO in the gas streams.

The following disadvantages presently indicate a lack of knowledge concerning the material rather than disadvantages in the material itself.

- (1) No information on CH_3I removal efficiencies or capacity.
- (2) The life cycle of regenerated AgZ material is not known.
- (3) No tests on extended temperature cycling have been made.
- (4) The effects of radiation on the material have not been examined.
- (5) Tests need to be performed to determine the effect of potential poisons to the filter bed.
- (6) Additional work is required to examine the proper storage medium for HI produced on regeneration.

5.5 Resins

The possibility of using resins to remove iodine has been examined by several researchers. Much of this work involved iodine removal from liquid streams; however, some limited studies were carried out in gas

streams. For the most part, this work was brought about to find iodine sorbents less expensive than silver impregnated ones or more stable than carbons.

Ion-exchanged resins are known for their adsorption and exchange properties. They have also been found to have good chemical stability. The macroreticular structure of the resins permit solutions to diffuse completely through the resin.

5.5.1 Chemical mechanism

Little work has been done to determine the mechanism by which elemental iodine or organic iodine reacts with the resins. In general, macroreticular resins exhibit attraction for substances due to their hydrophobicity or hydrophilicity. Since these resins are mainly nonionic in function, they show strong attraction for uncharged molecules such as elemental iodine or organic iodine, and they show only weak attractions for ionic iodide compounds.

5.5.2 Varion KS

In 1968, Hirling¹⁰⁴ reported on the efficiencies of various filter packings. Among the materials studied were Varion KS (a cation-exchange resin), silica gel, and polypropylene fibers. Tests showed at high loadings (875 mg I₂ to 1437 mg I₂/g of sorbent) the removal efficiencies were quite low (<40%). The beds at these loadings may have been saturated and not reflect the actual filter efficiencies. After silver impregnation, Varion KS removed HI with an efficiency of 93.1% at a loading of 1437 mg HI/g sorbent.

5.5.3 Amberlite resin

Moore and Howerton studied the applicability of the Amberlite series of sorbents from Rohm & Haas Company for iodine removal. The initial studies¹⁰⁵ indicated that Amberlite resins XAD-2 through XAD-12 were capable of producing DF's greater than 10⁴ for a loading of up to 213 mg I₂/g sorbent. These studies were conducted in dry air with an

airborne iodine concentration of 3×10^3 mg I_2/m^3 and $21^\circ C$. Later studies¹⁰⁶ with XAD-12 showed that there was some decrease in efficiency as the bed temperature was increased. Some of the iodine sorption capacity from XAD-12 was also lost in the presence of moisture. Decontamination factors of 10^3 to 10^4 were obtained for CH_3I removal using XAD-12 in dry air; loadings for CH_3I were low (10 to 84 mg CH_3I/g sorbent). Increasing temperature adversely affected CH_3I removal. Additional studies on polystyrene, polyurethane foam, and polypropylene were discontinued due to their very low capacity for elemental iodine.

XAD-12 was also compared to silver zeolite, iodized carbon, and Dowex 1-X4 (an anion exchange resin).^{107,108} The Dowex 1-X4 and AgX both showed lower capacities than the iodized carbon or XAD-12.

A final limited study by Moore¹⁰⁹ examined the effects of humidity on the capacity of XAD-4 and XAD-12. Decontamination factors greater than 10^3 were obtained at 60% relative humidity at $25^\circ C$ with loadings of 50 mg I_2/cm^3 of sorbent.

The effects of radiation exposure have been studied by several workers. Hingorani and Venkateswarlv^{110,111} looked at the effects on both Ag impregnated and unimpregnated Amberlite IR-120. Negligible radiation effects were noted on unimpregnated IR-120 up to 1 m rd; however, as the dose rate was increased to 17 m rd, the capacity decreased to 61% of the original amount. The silver impregnated sorbent showed no effect up to 20 m rd, which may be a result of surface oxidation of the IR-120 and the silver impregnation protection of the surface from such oxidation reactions. Huh et al.¹¹² examined the radiation-induced bonding of iodine to polystyrene. This work was done in aqueous suspensions with a population composed of 1.4×10^{-12} (0.234 μm in diameter) per cubic centimeter. Bonded to the surface are 0.02 iodine atoms per 100 eV of energy; this iodine was not removed by washing or exposure to anion exchange.

5.5.4 Process capacity

The maximum loading of elemental iodine on XAD-12 was 1.39 g I_2/g of sorbent at $25^\circ C$ from dry air; 84 mg CH_3I/g of sorbent was the maximum

loading of organic iodide on XAD-12 at 50°C from dry air. For XAD-4, the maximum loadings were 278 mg I₂/g and 15 mg CH₃I/g of sorbent from dry air at 21 to 25°C. The airborne iodine concentration was 1 to 4 × 10³ mg I/m³.

5.5.5 Advantages and disadvantages

Resins appear to offer advantages similar to those of other solid sorbents. In addition resins offer the following:

- (1) The macroreticular resins are acid resistant and thus may function well in gas streams containing NO_x.
- (2) High I₂ capacity.
- (3) Good resistance to radiation damage.

The limited amount of work with these materials also indicates the following disadvantages.

- (1) Adversely affected by humidity and elevated temperature.
- (2) Limited experience and information available on iodine removal from gas streams.

6. MISCELLANEOUS IODINE REMOVAL METHODS

In addition to the methods of iodine removal previously discussed, numerous other methods have been investigated. Some of these methods include the use of foams, sand and soil, and wall coatings. The tendency for iodine deposition on the walls of ducts and tubing was also studied. Iodine deposition of this kind is of concern, both as a contamination problem when deposition occurs on building surfaces and as a measurement problem in experimental tests where the uncontrolled iodine deposition in equipment lines could produce erroneous measurements of iodine flow.

6.1 Surface Deposition

Early work with iodine by Chamberlain¹¹³ showed iodine deposition on various surfaces which appeared to be controlled by aerodynamic factors at low Reynolds numbers. An analogy was drawn between the deposition of iodine and heat transfer by forced convection. Clean copper and filter paper were described as "perfect sinks" for iodine deposition. Other materials were tested to determine the relative amounts of iodine deposited compared to copper (see Table 12).

Since Chamberlain's report, numerous other investigators have examined iodine deposition. Davis et al.¹¹⁴ examined iodine deposition on structural surfaces, and Milstead et al.¹¹⁵ examined iodine deposition on low chromium-alloy steel. It was found that at 400°C, 3.8 to 23.2 µgI was deposited per cm². Hiller and Schad¹¹⁶ examined the following pipe materials to determine their retention of iodine: steel tubing — st 35, stainless steel tubing-4550, teflon tubing, polyethylene, and hard grade PVC. The plastic tubing was shielded with a grounded knit wire hose to reduce the buildup of static charges. The "pipe factors" (shown in Table 13) were determined based on a ratio of iodine in the outlet of the tubing to iodine in the inlet.

Experiments in Japan by Tachikawa et al.¹¹⁷ indicated that I₂ and HI are effectively trapped on copper metal; however, CH₃I must be converted into an inorganic form for effective trapping. No information on

Table 12. Iodine-131 deposition on flat plates of various materials relative to deposition on copper plate^a

Surface	Relative deposition to copper
Paper	0.9
Steel	1
Aluminum	0.5
Glass	0.3
Perspex	0.6
Copper with thin film of paraffin wax	0.04

^aFrom Ref. 113.

Table 13. Pipe factors for the retention of iodine on various pipe materials^a

Steel tubing - st 35	0.01-0.09
Stainless steel 4550	0.13-0.75
Teflon shielded	1.0
Polyethylene shielded	0.8
Hard grade PVC shielded	0.04-0.05
Teflon without shielding	0.66-0.68

^aFrom Ref. 114.

the process capacity is available. The copper system was regenerated by the use of high temperature H_2 as with AgX and AgZ. H_2 was also used to reduce the CuO to copper for effective trapping of iodine.

6.2 Soils

Other work in Japan focused on the use of sand and soil for iodine removal.^{118,119} Results indicated that 20 to 61% of the iodine initially adsorbed was removed by air sweeping of the bed. The quantity of iodine adsorbed decreased with increasing temperature and a more marked decrease in capacity was seen with increasing relative humidity. Loadings achieved vary from 30 to 350 mg I_2/g of sand.

6.3 Coatings

The application of coatings to retain iodine was examined at Battelle Memorial Laboratories.^{120,121} The major emphasis was on coatings to sorb iodine compounds onto the walls of the containment structure in the event of an accident involving a major release of fission products. In this work, advantage was taken of elemental iodine's formation of charge transfer complexes with amines. These complexes are stable at ambient temperatures and fairly stable at elevated temperatures. Quarternary ammonium salts and amine salts are formed by CH_3 and HI, respectively, with amines. The two most promising coating systems were found to be 1, 10-phenanthroline impregnated upon asbestos and the three component film of copolymer Genamid 2000 and Epon 828 as a binder with 1, 10-phenanthroline. At $37^\circ C$ 0.0065 mg CH_3I/cm^2 was adsorbed onto the phenanthroline on the asbestos system, and 0.0035 mg CH_3I/cm^2 was adsorbed on the Genamid, Epon, and phenanthroline coating.

6.4 Foams

Investigation of the application of foams to contain iodine¹²² was directed toward use in contained atmospheres in the event of a release of iodine. It was found that the addition of hydrazines to water provided high removal rates for elemental iodine. A 0.11 M concentration

of unsymmetrical dimethyl hydrazine in foam resulted in iodine reductions of 99% and 99.97% in 12 and 18 minutes, respectively. Foams containing 0.33 M tri-n-butylphosphine were found to require seven minutes to reduce CH_3I concentrations by 99%. The high removal rates of these foams were believed to be the result of gas-phase reactions with the bubble surface acting as a collecting surface for aerosol particles.

6.5 Potential Usefulness

With the exception of the deposition of iodine on the surface of materials, these methods have limited value in the treatment of off-gas from a reprocessing plant. These methods might, however, serve a role in emergency containment of iodine should an accident disable the in-line iodine removal system.

7. SUMMARY

This literature survey was conducted to provide information to evaluate the various methods that have been or are being developed to remove iodine from gas streams.

Iodine removal from off-gas streams is required to meet minimum standards for release levels as set forth in Appendix B of 10 CFR 20. These limits are 2×10^{-11} Ci/m³ for ¹²⁹I and 1×10^{-10} Ci/m³ for ¹³¹I.¹² However, when assessing the maximum acceptable limits, additional factors based on experimental evidence indicate that these limits should be lower. In a report from ORNL, the maximum acceptable airborne iodine limits from a LMFBR fuel reprocessing plant for ¹³¹I and ¹²⁹I are reported to be 4.8×10^{-14} Ci/m³ and 1×10^{-15} Ci/m³, respectively, at 2 to 3 km from the site. To achieve these levels, decontamination factors of 3×10^6 and 1×10^3 are required for a 5-t/d plant with fuel cooled 60 or 150 days, respectively.¹³

During the past two decades, various systems have been studied to reduce the iodine released to the environment. Two liquid systems are available: the nitric acid (Iodox) system and the mercuric nitrate - nitric acid (Mercurex) system. Solid sorbents have also been studied, either as secondary systems to provide final filtering after either Iodox or Mercurex processes or as primary systems to replace the liquid methods. These solid sorbents include silver zeolite, silver mordenite, alumina-silicates, and macroreticular resins. Activated carbon was also examined as a reference material.

The Iodox process employs hyperazeotropic nitric acid in a bubble cap tower to remove iodine species from a gas stream. Tests show DF's up to 10^5 and the ability to handle feed concentrations of 0.02 to 200 mg I₂/m³. The system has also been shown to be capable of maintaining a DF greater than 10^4 in the presence of high concentrations of H₂O and NO₂ (2.5% and 7.8%), while operating at ambient temperatures. Iodobenzene and benzene contaminants appeared to have a minimal effect on the system performance, although the amount of these aromatic compounds that was introduced into the system is in question. The removal efficiency

is not too sensitive to liquid flow as long as the acid concentration is maintained above 20 M HNO_3 and iodine solubilities are not exceeded. A possible disadvantage of this process is the complexity of the system which includes not only the Iodex column but also an extractive distillation column employing $\text{Mg}(\text{NO}_3)_2$ to produce the hyperazeotropic nitric acid and an evaporation system to produce the solid product containing the iodine. A second disadvantage is the use of highly concentrated nitric acid which necessitates the use of zirconium or titanium materials of construction.

The Mecurex process yields DF's greater than 10^3 for both CH_3I and I_2 at ambient temperatures from off-gases containing high concentrations of NO_x . The NO_x improves the process efficiency by reducing iodate formation; however, aromatics severely reduce the DF's from 10^4 to 10^2 .

Activated carbon, while used extensively in nuclear power plant ventilation and confinement systems, cannot be considered as a prime candidate for the removal of iodine from fuel reprocessing plants for the following reasons: (1) it has a low ignition temperature, (2) it is adversely affected by nitrogen oxide, which could also form explosive compounds, and (3) it has a high iodine desorption rate at an elevated temperature.

Silver zeolites (AgX) are capable of adsorbing iodine up to 500°C and retaining it up to 1000°C . Water vapor has been shown to degrade the removal efficiency of AgX; this effect was more pronounced for the removal of CH_3I than I_2 . Decontamination factors for I_2 removal at 90% relative humidity at 25°C remain at 10^4 , nearly that of dry air removal. For CH_3I , removal of DF's are reduced to about 20, which can be overcome by heating the bed to a temperature of 100 to 200°C . To make the use of silver zeolite more cost effective, work has been done on regenerating loaded AgX with H_2 at 500°C . However, a 50% reduction in capacity was noted after five cycles. Silver zeolite, as other solid sorbents, has the advantage that the iodine loaded form is a solid suitable for long term storage. Silver zeolite is not affected by most organic or inorganic contaminants; the exceptions are dodecane, H_2S , SO_2 , Cl , and to a lesser degree C_3H_8 and NO_2 .

Decontamination factors in excess of about 10^3 have been obtained using GX-135, a granular, silver salt containing aluminum and silicon oxides from North American Carbon Company, with loadings of $10.9 \text{ mg CH}_3\text{I/cm}^3$ at 200°C . This sorbent shows the same trend as other sorbents with respect to relative humidity, i.e., a noticeable decrease in efficiency with increased relative humidity. Work in West Germany on another silver containing alumina silicate, AC-6120, shows similar results. The silver content of AC-6120 is significantly less than that of AgX, 70 to 120 mg Ag/g sorbent versus 360 mg Ag/g sorbent for AgX; this should lower its initial cost. Reports also indicate that NO_2 in the off-gas stream is beneficial to iodine removal by preventing the oxidation of the silver.

Silver-exchanged mordenite performs similarly to silver zeolite, however, the substrate is much more acid resistant as indicated by a higher $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio. This material also shows better recycle potential than AgX. After 13 recycles, the material retained 80% of its original capacity. Tests thus far have been limited to elemental iodine, and little work has been done to show the effects of potential contaminants. Tests with nitrogen oxides indicate that 2% NO in the gas stream will negate the adverse effects of 6% water and/or 2% NO_2 .

Only limited studies have been carried out on the iodine removal characteristics of macroreticular resins from gas streams. The loading capacity of Amberlite XAD-12 (a polymeric adsorbent from Rohm & Haas Company) was high ($1.39 \text{ gI}_2/\text{g}$ sorbent at 25°C from dry air and $84 \text{ mg CH}_3\text{I/g}$ at 50°C from dry air). Corresponding DF's were 10^2 to 10^3 for I_2 removal and 10^4 for CH_3I removal. However, the removal efficiencies of these sorbents are seriously affected by increases in relative humidity, e.g., at 25°C and 60% relative humidity, the capacity of XAD-12 is only $60 \text{ mg I}_2/\text{g}$ sorbent. This material was also adversely affected by temperatures in excess of 50°C .

Other removal methods (sand, soil, various coating, and foams) were briefly examined, although they showed little promise for the nuclear reprocessing industry.

The Iodox system is presently the most promising system for the primary iodine removal system. It has already been tested on an engineering scale and has demonstrated good capabilities and operability. There are, however, concerns about the long-term integrity of the piping in contact with the 20 to 23 M HNO₃. Silver mordenite appears to be the most promising of the solid sorbents developed to date; however, this material will require additional testing with CH₃I and engineering scale tests of filter beds and regeneration facilities. AC-6120 also shows potential on a once-through basis and is commercially available at the present time.

APPENDIX A

Physical Constants of Methyl Iodide

Formula	CH ₃ I
Alternate names	Iodomethane
Molecular weight	141.95
Form and color	Colorless liquid
Specific gravity at 20°C relative to 4°C	2.279
Freezing point, °C	-64.4
Boiling point, °C	42.4
Critical temperature, °C	255
Critical pressure, atm	54.6
Solubility, g/100 g of water	
At 20°C	1.42
At 22°C	1.38
Ethanol	Infinite
Ether	Infinite
Surface tension at 20°C, dynes/cm	30.14
Viscosity at 20°C, Cp	0.46

Vapor pressure

$$\log_{10} P_{\text{mm}} = -\frac{1475}{T} + 7.56$$

T in K

Heat capacity for range 298.1 to 600 K

$$C_p^\circ = 4.105 + 24.487 \times 10^{-3}T - 9.733 \times 10^{-6}T^2$$

For T in K

$$C_p^\circ = \text{cal mole}^{-1} \text{K}^{-1}$$

APPENDIX B

Relationships Between Methods of Reporting Iodine Removal

$$\text{Removal Efficiency (\%)} = \left[\frac{\text{inlet concentration} - \text{outlet concentration}}{\text{inlet concentration}} \right] (100)$$

(RE)

$$\text{Decontamination Factor} = \frac{\text{inlet concentration}}{\text{outlet concentration}}$$

(DF)

$$= \frac{100}{100 - \text{RE}}$$

Decontamination Efficiency = Removal efficiency

$$\text{Percent removal} = 1 - \left(\frac{1}{\text{DF}} \right) 100$$

Reactor equivalent delay (RED) is the reduction in concentration as compared to the reduction in concentration due to normal decay.

$$\text{DF} = 2^{\text{RED}/t_{1/2}}$$

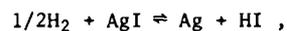
where $t_{1/2}$ is the half-life of the isotope.

For example, a DF of 4 for ^{131}I removal would be the result in a RED of 16.1 or twice the half-life of ^{131}I .

APPENDIX C

Thermodynamic and equilibrium data for HI during regeneration of AgX filter beds with H₂

For the reaction



the calculated thermodynamics and kinetic data are:^a

T (°C)	ΔH (cal/mole)	ΔS (eu) ^b	ΔG (cal/mole)	K _p P _{HI} /P _{I₂} ^{1/2}	P _{HI} (atm)				
					0.05 atm H ₂	0.1 atm H ₂	0.25 atm H ₂	0.5 atm H ₂	1.0 atm H ₂
400	1.8045 × 10 ⁴	9.8415	1.1421 × 10 ⁴	1.9579 × 10 ⁻⁴	4.3780 × 10 ⁻⁵	6.1914 × 10 ⁻⁵	9.7894 × 10 ⁻⁵	1.3844 × 10 ⁻⁴	1.9579 × 10 ⁻⁴
450	1.7875 × 10 ⁴	9.6468	1.0899 × 10 ⁴	5.0784 × 10 ⁻⁴	1.1356 × 10 ⁻⁵	1.6059 × 10 ⁻⁴	2.5392 × 10 ⁻⁴	3.5910 × 10 ⁻⁴	5.0784 × 10 ⁻⁴
500	1.7712 × 10 ⁴	9.4922	1.0373 × 10 ⁴	1.1680 × 10 ⁻³	2.6117 × 10 ⁻⁴	3.6935 × 10 ⁻⁴	5.8399 × 10 ⁻⁴	8.2589 × 10 ⁻⁴	1.1680 × 10 ⁻³
550	1.7556 × 10 ⁴	9.3779	9.8370 × 10 ³	2.4435 × 10 ⁻³	5.4638 × 10 ⁻⁴	7.7270 × 10 ⁻⁴	1.2217 × 10 ⁻³	1.7278 × 10 ⁻³	2.4434 × 10 ⁻³
600	1.7408 × 10 ⁴	9.3038	9.2841 × 10 ³	4.7424 × 10 ⁻³	1.0604 × 10 ⁻³	1.4997 × 10 ⁻³	2.3712 × 10 ⁻³	3.3534 × 10 ⁻³	4.7424 × 10 ⁻³
650	1.7266 × 10 ⁴	9.2700	8.7086 × 10 ³	8.6719 × 10 ⁻³	1.9391 × 10 ⁻³	2.7423 × 10 ⁻³	4.3360 × 10 ⁻³	6.1320 × 10 ⁻³	8.6719 × 10 ⁻³
700	1.7132 × 10 ⁴	9.2764	8.1047 × 10 ³	1.5125 × 10 ⁻²	3.3820 × 10 ⁻³	4.7829 × 10 ⁻³	7.5624 × 10 ⁻³	1.0695 × 10 ⁻²	1.5125 × 10 ⁻²

The equilibrium constant may be calculated by

$$K_p = \left(\frac{P_{\text{HI}}}{P_{\text{H}_2}} \right)^{1/2} = 208.22 e^{-9343/T}$$

with T = K .

^aFrom Ref. 92.

^bEntropy units.

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