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THE CHEMICAL BEHAVIOR OF IODINE IN AQUEOUS SOLUTIONS UP TO 150°C: AN INTERIM REPORT

L. M. Toth O. L. Kirkland

ABSTRACT

The chemistry of iodine, I_2 , has been examined in (pH = 6 to 10) aqueous solutions containing 2500 ppm B as H₃BO₃ (0.231 M) at temperatures up to 150°C using absorption spectrophotometry to identify and monitor the iodine species present. Three objectives have been considered: (1) species identification with special attention given to HOI; (2) the kinetics of reaction between iodine and water to produce iodide and iodate ions; and (3) partition coefficients between liquid and vapor phases for individual iodine species.

Because this report is interim in nature, none of the above topics have yet been completed. Nevertheless, the current status is described herein. No spectral evidence has been found for HOI in the vapor even though an ultraviolet absorption band is predicted. Kinetics for the second stage of the iodine-water reaction, i.e., the disproportionation of HOI, have been studied at 65°C and found to vary from minutes at the high pH values to hours at the lower values. Partition coefficients for I_2 have been measured and found to deviate only slightly (by ~5%) from those predicted using Henry's Law.

INTRODUCTION

The overall objective of this study is to determine the various iodine species present in the primary system and containment conditions representative of hypothetical accidents in PWRs and BWRs. The immediate goals of the study are then (1) to identify the iodine-bearing species that would be present is a range of LWR conditions and (2) to determine the effective volatility (i.e., partition coefficients) for these conditions. Since little published fundamental data related to aqueous iodine chemistry treat conditions greater than 75°C, the current interests have been directed at obtaining these data at higher temperatures. The present experimental limits have been confined to <150°C for the purpose of defining (1) the species present, with special attention given to the characterization of the elusive HOI entity; (2) the kinetics of the iodine reaction with water containing pH-adjusted boric acid; and (3) the partition coefficients for each of the identified species.

This present study has focused on the hydrolysis reactions of molecular iodine, I₂, under nonredox conditions because so much discussion has been given to speculations on the chemical behavior of

I₂ following its assumed release during a reactor accident. The major parameters that were examined were temperature, pH, and iodine concentration for an aqueous solution containing 2500 ppm B as boric acid, H₃BO₃ (i.e., 0.231 M). The pH of the boric acid solution was varied between 6 and 10 in unit increments by NaOH additions. The sensitivity of the spectrophotometric method used here has permitted analyses for iodine at concentrations down to 1×10^{-6} M and the range considered here has been [I₂] = 10^{-3} to 10^{-6} M.

This interim report presents the results obtained to date since the inception of work nine months ago.

EXPERIMENTAL

Molecular iodine tends to react very readily with most common metals to form metal iodides. When the intent of a particular study is to examine iodine reactions with media that do not include the container vessel, precautions must be taken to insure that the container is either protected from or unreactive towards the iodine medium. Considerable attention had to first be devoted to the development of a suitable container prior to the actual study of the aqueous iodine chemistry; two satisfactory containers that are useful up to 150°C have been selected and are described later in this section. Nevertheless, the ultimate intent of this study is to examine the aqueous chemistry of iodine at temperatures up to 304°C; and for these purposes, much more sophisticated containers will be necessary. If this temperature goal remains in effect, then further container development and testing will be a necessary prequisite prior to proceeding above 150°C.

Pyrex glass is by far the most frequently used container material for laboratory scale experiments involving aqueous iodine solutions because reactivity with glass is not observed under ordinary (ambient) conditions. However, the usage of glass systems has usually been limited to temperatures less than 100°C due to the increasingly high water vapor pressures that must be contained. Consequently, the most practical systems for containing aqueous iodine solutions at temperatures in excess of the boiling point of vater would appear to be unreactive metallic containers. The development, design, and construction of a suitable container system for the aqueous iodine studies up to 150°C has therefore proceeded along the consideration of the following sequence; stainless steel vessels; plastic-coated or gold-plated metal vessels; titanium vessels; and ballast-pressured glass cells. Characteristics of each of these are presented below.

<u>Stainless steel</u>. Type 304L, like all iron alloys, is unsuitable for studies of iodine solutions due to rapid corrosive loss of I_2 from solution, even at ambient temperatures. Steel cells are, however, useful as secondary containers for the ballast-pressured glass cells (see below).

Plastic-coated metals. Teflon or other halocarbons can be readily applied to metal surfaces by commercially established coating techniques. However, the halogens are significantly soluble in these materials and tend to diffuse through them. The slow loss of iodine by such a diffusion mechanism might be acceptable if it were not for the corrosion of the metal underneath and subsequent loosening of the coating from its attachment to the metal surface. High temperatures promote this diffusion to such an extent that 100°C appears to be the useful limit for coated cells in contact with iodine solutions; but simple glass cells are also useful up to this temperature, so no advantage is seen in using plastic coatings. Two final properties of these coatings should be noted: First, they are most successfully attached to metal surfaces by first applying a primer coat that contains metallic elements such as cobalt; and if the primer is not thoroughly covered by the final coat, it provides a very reactive surface for I2 attack. Second, the plastic materials have very high thermal coefficients of expansion that can cause thick coatings to buckle and peel off the metal substrate.

<u>Gold-plated metals</u>. Even though corrosion reactions are eliminated by such coats, direct halogenation of the gold appears to be responsible for the continuous and possible reversible loss of iodine from aqueous solution. Noble metal coats are therefore not considered to be practical for long term usage in contact with aqueous solutions under the conditions of these experiments.

<u>Titanium containers</u>. At pH > 5, corrosive loss of I₂ is either eliminated or too slow to be of consequence. Therefore, we believe that titanium is a suitable material for kinetic studies. The availability of a titanium optical cell in this laboratory has further encouraged its usage; and stoichiometry tests on the I₂ hydrolysis reaction have been used (see Results and Discussion) to demonstrate that the homogeneous aqueous phase reactions are the dominant chemical processes taking place.

Ballast-pressured glass cells. The low structural integrity of silica can be circumvented by balancing the vapor pressure of water on the inside of a silica cell with an equivalent pressure of gas on the outside. A custom design silica cell, Fig. 1, has been constructed. It is normally filled with an aqueous solution containing iodine and other reagents and the filling tubes then sealed off with a torch. The cell is next loaded into a heavy-walled steel cell which can be pressurized with dry helium so that there is a minimal pressure drop across the glass cell. The pressure of helium that is applied to the exterior of the cell corresponds to the vapor pressure of the water within the cell. This cell has been used at 150°C on many occasions; and it is believed that even higher temperatures could be readily achieved.

As a result of the developmental work, two basic containment systems were selected for the experiments described in this report. The first was a titanium cell (Fig. 2) with optical pathlength of 25.1 cm that was normally filled above the sapphire optical windows with ~400 mL of aqueous boric acid solution that had been pH-adjusted with NaOH

Fig. 1. Photograph of two chamber ballast-pressured silica cells. Small dam can be seen inside large chamber at point where right connecting tube is attached.

ORNL-Photo 0792-81

ORNL-Photo 6159-81



Fig. 2. Exploded view of titanium cell. Two 5-in.-diam band heaters can be seen around the body of the cylindrical cell-

solution. A small glass capsule containing purified iodine was placed in the cell during assembly and a titanium breaker rod poised above it so that once the system had equilibrated at a particular temperature, a sharp blow to the exterior end of the rod would cause sudden release of the I₂ into the aqueous medium. For additions to the aqueous medium at temperatures <100°C, aqueous iodine solutions of low pH were loaded in the capsules. Then, when the capsule was broken the previously dissolved iodine solution could simply mix with the boric acid buffer solution. On other occasions (namely, at 65°C), an aqueous stock solution of I₂ in distilled water was admitted through a separatory funnel attached to the top of the titanium cell. No differences were observed due to the method of addition as long as the contents of the cell were thoroughly mixed after the addition of the iodine solution.

The second system used in this work was a ballast-pressured silica cell (Fig. 1) consisting of two cylindrical chambers connected by silica tubing. A small dam in the larger chamber promoted the pumping of solution around through the smaller chamber when the system was rocked. The 19 mL solution volume completely filled the small liquid chamber and connecting tubes leaving just enough liquid in the larger, vapor phase chamber to slosh over the dam when the assembly was rocked. Gentle rocking of the cell insured equilibration of liquid and vapor phases by circulating the solution through the two chambers. Absorption spectra of either liquid or vapor phases could be measured by merely shifting the cell assembly up or down so that the appropriate window intersected the spectrometer light beam. The liquid and vapor pathlengths of this cell were 2.08 and 30.3 cm, respectively. The system was best suited for long-term equilibration experiments where distribution coefficients of iodine and species absorption spectra were sought.

Absorption spectra were monitored with a special Cary 14 recording spectrophotometer that had an enlarged sample-reference compartment area. A cradle in the compartment permitted the rocking of large sample cells between spectral measurements. Because the response time of the spectrometer was of the order of 1 s, it was not possible to monitor very fast kinetic processes with this instrumentation. Furthermore, the introduction or release of reagents into a previously equilibrated aqueous solution, followed by mixing, usually required up to 1 min before spectral measurements could begin. Therefore, most well-mixed solutions were not monitored until 1 min after the reagents had been added. In some instances, however, unmixed solutions were monitored throughout the introduction of reagents and here the 1 s response time of the instrumentation governed the time limitations of the measurements. The results from these unmixed solutions vary greatly from those of the homogeneous solutions, mainly because of local concentration that altered the hydrolysis rate of iodine. These unmixed solutions have been studied briefly by high speed motion picture photography and the results will be contrasted with those of the homogeneously mixed solutions.

The wavelength range of the spectrophotometer extends from 2500 to 190 nm; but with the interferring absorption bands from water overtones in the near infrared region, the practical spectral range for aqueous solutions covers the 800 to 190 nm region. However, when the gas space above the aqueous solutions is examined, it is often possible to cover the full spectral region of the instrument from the near infrared limit to the ultraviolet. Iodine species typically absorb in the visible-uv region with molar absorptivities that often permit detection of some at quite low concentrations. Typical spectra of these iodine species are shown in Figs. 3 through 8 for condensed as well as volatile forms. Table 1 gives the wavelengths and molar absorptivities of the various species that were monitored.

Species		ε	(λ)	Detection limit (×10 ⁻⁶)	
Liquid	I ₂	642 17	(460) (270) (205)	2.0	
	I3-	26,400	(350)	0.6	
	I	13,500	(225)	0.12	
	103-	5,332	(200)	0.3	
	CH3I	173	(248)	9.6	
Vapor	I ₂	831	(533)	2.0	
	HI	356	(258)	4.7	
	CH3I	2,833	(201)	0.6	

Table 1. Iodine species, molar absorptivities (ε) and detection limits currently identified

 ε units in M⁻¹ cm⁻¹, wavelength λ in nanometers.

Molarity, as determined from Beer's Law, $c = A/\epsilon l$, where A = 0.05, and l = 30 cm for this system and ϵ was taken from the preceeding column.

RESULTS AND DISCUSSION

When molecular iodine is released into water, it is generally accepted (1) that it reacts according to the following equilibria:

$$I_2 + H_2O = HOI + I^- + H^+$$
, (1)
 $3HOI = IO_3^- + 2I^- + 3H^+$. (2)



Fig. 3. Visible absorption spectrum on I_2 vapor at room temperature.



Fig. 4. Ultraviolet absorption spectrum of weak iodine band at 270 nm.







Fig. 6. I_3 "desorption spectrum in aqueous solution at 20°C. The sharp ultraviolet cutoff, however, is due to I which was added in excess to convert all the I_2 reagent to I_3 .

H



Fig. 7. Relative absorption spectra of 1⁻ and IO_3^- in aqueous solution at 20°C. Molar absorptivity of 1⁻ peak at 225 nm is 13,500 M⁻¹ cm⁻¹.



For the purpose of the following discussion, we shall define Eq. (1) as the first stage and Eq. (2) as the second stage of the iodine reaction with water. It is also generally recognized (1) that the first stage reaction is very fast and usually reaches equilibrium conditions within 1 s, whereas the second stage is much slower and takes anywhere from seconds to years (depending on temperature, pH, etc.) to reach equilibrium. These two reactions are often combined and presented in terms of the overall reaction:

$$3I_2 + 3H_20 = IO_3^- + 5I^- + 6H^+$$
 (3)

The stoichiometry of the overall reaction provides a useful test by which this reaction can be distinguished from a corrosion related reaction where all of the iodine is expected to be converted to I^- .

Because so much discussion of aqueous iodine chemistry has previously revolved about the poorly characterized HOI intermediate species (for example, see ref. 1 and references contained therein), it was considered imperative that much of the initial effort should be devoted to the detection and characterization of HOI. This problem was therefore considered prior to kinetic and partition coefficient measurements.

HOI Characterization

Although HOI or the OI⁻ ion have not been directly identified under conditions similar to those found in LWR and BWR aqueous systems, a brief examination of the recent chemical literature leads one to the conclusion that the hypoiodite ion, OI⁻, has been fairly well established. This conclusion arises mainly from a comparison of the results of A. Treinin et al. with those known for the OCI⁻ and OBr⁻ analogs.

Treinin first reports³ the electronic spectrum of OI⁻ which is produced by dissolving iodine in 4 N NaOH to make solutions that are $\sim 10^{-2}$ M in OI⁻. He also measured a second order rate of disappearance for the absorption bands attributed to OI⁻ (at 365 and ~ 270 nm) which is given as 4 × 10⁻² M⁻¹ s⁻¹, i.e., a solution of 0.04 M IO⁻ decomposed within 80 min. The reaction is reported to be a disproportionation:

 $OI^{-} + OI^{-} - OI_{2}^{-} + I^{-}$,

and is claimed to be analogous to the OC1⁻ and OBr⁻ disproportionation reactions. Treinin later gave more details on the disproportionation⁴ and the photolysis⁵ of OI⁻.

Comparison of Treinin's spectrum at 365 nm with those reported 6 for OC1⁻⁻ and OBr⁻⁻ lead to some interesting correlations:

Species	Band	Molar absorptivity
0C1-	290 nm	360
OBr ⁻	331	326
01-	365	32

The only anomaly rests on the unusually low value for the OI molar absorptivity.

The corresponding values reported⁶ for the hypohalous acids are:

?

HOC1	240	90
HOBr	260	92

from which we could estimate that HOI would occur at:

HOI 280

Since the equilibrium constant for the dissociation of HOI at 25°C is reported to be 2.3×10^{-11} , ⁷ the OI⁻ ion should not be a significant species at pH < 10. Therefore, attempts to characterize the HOI intermediate under simulated reactor accident conditions should be on the undissociated form; and with so many known aqueous iodine species absorbing strongly in the ultraviolet region (e.g., I⁻, IO₃⁻, and I₃⁻), the greatest chances of isolating an HOI spectrum occur in the vapor region where only I₂ and, possibly, HI could be present.

We have therefore examined the vapor phase over an aqueous solution contained in the ballast-pressured silica cell at temperatures up to 150° C with the intention of identifying any bands that could be attributed to an HOI species. It was anticipated that this species might be volatile enough at this temperature to be present in concentrations that could be detected by absorption spectrophotometry. However, no bands other than those assigned to I₂ have been observed in the vapor. Furthermore, by digitally subtracting sample and reference spectra, the region around the very weak 270 nm I₂ band (see Fig. 4) has been shown to contain no unresolved bands that might be attributed to species other than I₂. In an alternate approach, we have studied the vapor equilibrium,

 $H_2O + I_2 = HI + HOI$,

by using such small amounts of I_2 and H_2O in a sealed silica cell that the equilibrium vapor pressure of neither reagent was attained at 150°C. In this procedure, then, there was no liquid phase into which the HI would dissolve. Examination of the vapor absorption spectrum revealed only I_2 — and no HI as might be expected from the above equilibrium. Using the measured absorptivity for HI in the vapor, we have calculated an upper limit for the equilibrium quotient of the <u>vapor</u> reaction at 150°C, $Q = [HI][HOI]/[I_2] = 10^{-8}$. Therefore, we have no direct spectral evidence for an HOI species at the present time. However, if it does indeed have an absorption band in the ultraviolet as predicted, we still have some hope of detecting it in solution by digital analyses of the data.

Kinetic Studies

The rate at which iodine reacts with water via Eqs. (1) and (2) determines the extent to which I_2 vapor or intermediates such as HOI can distribute to the vapor phase. Although kinetic data exist for these reactions at near ambient temperatures, there are little if any data at temperatures of 100°C and above. As part of the objectives of this work, we are seeking to establish data for these reactions that will permit a firm understanding of the lifetime of iodine species both in solution and in the vapor phase in contact with these solutions.

Several basic points concerning the nature of this experimental work should first be emphasized. In following the progress of this kinetic process, the iodine reactant, the intermediates, or the product ions could, in principle, be monitored. As indicated in the preceeding section, we have not yet been able to detect the "HOI"* intermediate as has been done for the OIT ion in 4 M NaOH solutions.4 Furthermore, the 103 species occurs at the ultraviolet limit of detection for our instrumentation and will often be obscured by the extremely intense absorption of I; and the I2 reactant either disappears within the response time of the spectrometer for high pH's or otherwise is often not present in concentrations that are great enough to be routinely monitored. With these considerations in mind, the greatest information at the present stage of this work can be obtained by monitoring the rate of growth of the I band occurring at 225 nm (see Fig. 8). The rate of formation of I typically follows the trend illustrated in Fig. 9. Immediately after release of iodine into the boric acid buifered solution in the pH = 7 to 10 range, the iodine hydrolyzes within 1 s via Eq. (1) to form I in concentration equal to that of the initial iodine. We must assume for the present that an equivalent amount of intermediate is formed according to this first stage reaction since its presence has not been verified under the conditions of these experiments. What one observes when monitoring the I" band is an immediate increase in absorbance to the point marked "a" as shown in the figure. Thereupon, a further gradual increase in the intensity of the I" band occurs [which is believed to be due to the second stage mechanism of Eq. (2)] until the final concentration of IT is equal to 5/6 that of the original gram atoms of iodine added. [Note that the stoichiometry of Eq. (3) predicts this.]

The stoichiometry test is then a useful means of demonstrating that a disproportionation mechanism like that of Eq. (3) is occurring as opposed to a corrosion process involving the titanium cell which is expected to convert all of the iodine to I^- .

*Given in guotes to emphasize assumed and not yet identified.

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Fig. 9. Growth of the I⁻ band for a typical kinetic experiment. Conditions for the curve shown here were 2×10^{-6} M I₂ in pH = 8, 0.231 M H₃BO₃ solution at 65°C.

The first attempts at measuring rates of reaction were made at 150°C, but inconsistencies in the data occurred due to the speed of the overall reaction and our failure to mix the solution adequately after release of the iodine. It was therefore reasonable to examine lower temperatures and gain experience that could lead to an improved procedure at the higher temperatures; the 65°C temperature was selected because it represented one point of interest in our program guidelines. The results of the kinetics study at various pH values are given in Table 2.

				pН		
[12]	6		7	8	9	10
10 ⁻¹⁴ <u>M</u>	25% at 68	h	>2 h	1.2 h	0.9 h	0.75 h
2×10^{-6}	1 >10 h		16 h	4 h	1.9 h	1.4 h

Table 2. Iodine kinetic study at 65°C as a function of pH and iodine concentration. The times given in the table are for the second stage reaction to go to completion, except in the case for 10^{-4} M at pH = 6, which had not reached completion by the indicated time.

The times for the reaction to reach completion for the 2×10^{-6} M I₂ at pH = 9 and 10 appear to be unreasonably similar and will be considered further in more detail. In addition, it was sometimes noted in following the rate of change in the I⁻ band that the band reached a maximum and then decreased over a period of time equavalent to that required to reach the maximum value given in the table. We have not included these data because they are still being examined. If they do prove to be consistent, they would indicate the presence of an intermediate that has an absorption band overlapping that of I⁻.

Finally, a graphic perpherial analysis was performed during the early stages of the 150° C kinetic studies in order to assess the validity of the experimental observations. It involved the use of high speed (500 frames per second) motion picture photography to record the events immediately following the release of 10 mg molecular iodine into 365 cc of boric acid solution at 150° C and pH = 7. Although the iodine was released into an unstirred environment with, presumably, a great localized decrease in the pH of the solution, the amber color of the molecular iodine species disappeared within 3 s. Direct observations such as these confirm the two-stage mechanism of Eqs. (1) and (2) in which iodine is first rapidly converted into less volatile products.

Distribution Coefficients

Since I₂ is the only iodine species presently found in both the liquid and vapor phase, it is the only species for which partition coefficients have been sought at this time. In order to simplify the presentation of the data for future use, we will report partition coefficients for individual species, i.e., D = (molar concentration in the liquid phase)/(molar concentration in the vapor phase). These are far more practical to produce and analyze prior to their assembly into the recently discussed composite coefficients (see ref. 1, Eq. 5.7).

The partition coefficients for iodine as a function of temperature are shown in Table 3 as determined after equilibration in the ballastpressured silica cell. Also shown in the table are those coefficients that would be predicted using available Henry's Law constants. (These constants, k, can be derived for various temperatures from reported solubility and vapor pressure data. Thus, $k = P_B/X_B$ where P_B is the vapor pressure of the pure solute and X_B is its mole fraction at saturation for a given temperature.) In all the cases shown, the actual partition coefficients are slightly greater than the predicted values due to the lower concentration of I_2 in the vapor phase. The solvation of iodine by the water is believed to be responsible for the decreased vapor pressure of the iodine over iodine-containing solutions and this association should decrease with temperature as indicated by the trend in the differences between the measured and predicted partition coefficients.

Temperature	Distribution coefficient		
(-c)	Measured	Predicted	
19	108	97.1	
52	29.7	27.2	
86	11.6	11.4	
92	10.9	10.3	
151	5.1	5.2	

Table 3. Iodine partition coefficients for 19 to 150°C in silica cells containing only I₂ in water (i.e., no buffers or pH adjustments)

CONCLUSIONS

Although this work is still in progress, certain observations can be drawn about the chemical behavior of molecular iodine when it contacts relatively high pH solutions (pH = 7 to 10) at moderately high temperatures. It is clearly evident that the first stage reaction is very rapid; and, as a result, we are only able to kinetically examine the second stage reaction involving the slower conversion of the presumed HOI intermediate into iodide and iodate ions. Nevertheless, this second stage reaction is very dependent on the particular pH, temperature, and concentrations chosen. At 65° C, the second stage reaction can go to completion in less than an hour at a pH = 10, while at higher temperatures much faster rates are anticipated.

The HOI intermediate has not yet been observed in spite of predictions concerning its detectability. Especially noteworthy is its absence in the vapor above equilibrated mixtures of iodine in water at 150° C. As a result of these data, it is evident that the equilibrium constant for the vapor phase reaction of iodine with water to form HOI and HI is less than 1×10^{-8} .

Finally, measurements of $\rm I_2$ partition coefficients up to 150°C show only a slight (<5%) positive deviation from Henry's Law which is explained by the solvation of iodine by water molecules.

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