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A Division of North American Aviation, Inc.

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ATOMICS INTERNATIONAL <small>A Division of North American Aviation, Inc</small>		NAA-SR- MEMO TDR NO 9507		APPROVALS	
TECHNICAL DATA RECORD		PAGE 1 OF 35		<i>Louis Bernath</i>	
AUTHOR G. O. Haroldsen <i>GH</i>		DEPT & GROUP NO 721-14		DATE 3/12/64	
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TITLE Hermetic Sealing Problems in a Small Boiling Water Reactor		S/A NO 1211		TWR	
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PROGRAM SNAP 4		SUBACCOUNT TITLE Subsystem & Component Cycle & Performance Analysis		AUTHORIZED CLASSIFIER SIGNATURE <i>Louis Bernath</i>	
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STATEMENT OF PROBLEM Substantiate the basis of the estimates of the rate of radiolytic decomposition in SNAP 4; size the recombiner, and validate its proper function in a hermetically sealed system.					
ABSTRACT The results of an extensive survey of the literature and of the subsequent analysis permit the prediction of the rate of radiolytic decomposition of the water coolant. Due to the difficulty in assessing the rate of the back reactions, fundamental data from water radiation chemistry could not be applied in a straightforward manner. It proved more expedient to employ the actually observed decomposition rate data from other boiling water reactors and to extrapolate to the operating conditions of SNAP 4. By this approach, the water decomposition rate was estimated to be 5 lbs/day. To assure adequate capacity under all conditions, the catalyst bed was sized to handle twice this rate. Based on the design method of Garber and Peebles ⁽²³⁾ , a catalyst bed 3 inches in diameter by 12 inches long, consisting of platinum-coated alumina pellets, appears ample. Operation of the recombiner in a closed recirculatory system was analyzed. It appears that an excess of hydrogen will result as oxygen is preferentially consumed in the initial corrosion process (metal oxidation). Once a stable oxide film has been established on the stainless steel surfaces and the excess hydrogen has been drawn from the system, the recombiner can thereafter handle the radiolytic gases satisfactorily.					

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I. INTRODUCTION

Although the radiolytic decomposition of water into hydrogen and oxygen as it will occur in SNAP 4 reactors will be rather small, of the order of a few pounds per day, it has nevertheless been a problem of considerable concern. The estimated radiolytic rate has repeatedly come under question, and the complications and magnitude of the gas handling problem have often been misunderstood. It seems appropriate to present in a single document all the information that has been used in estimating the decomposition rate and all the information dealing with the proposed radiolytic gas recombiner design. This report is intended to satisfy this need.

With regard to the radiolytic decomposition rate, first the literature was searched for all appropriate information. From the body of information collected, the many factors affecting water radiolysis were evaluated as to their specific contribution to the SNAP 4 decomposition rate. A best estimate was then made, along with a few supporting estimates based on certain particular conditions which could possibly exist. The latter served in identifying the maximum limit.

Next, the catalyst volume of the recombiner was estimated for a number of selected conditions of flowrate, temperature, and gas stream composition. Besides estimating the maximum volume of catalyst that conceivably would be required, the sensitivity of the recombiner to changes in flowrate, temperature and gas stream composition was also brought out. The largest volume requirement became the basis for the recombiner design. Since one of Atomics International's ultimate goals in the development of a hermetically sealed system, recombiner performance was evaluated as to how it would contribute to and be affected by hermetic sealing requirements. Of particular importance was a study determining the quantity of uncombined gases that might accumulate in a recycling system.

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II. CONCERNING RADIOLYSIS OF WATER

In his book on the radiation chemistry of water,⁽¹⁾ A. O. Allen states, "Of all the radiation chemical reactions that have been studied in aqueous solution, the most complex and bewildering is the decomposition of water itself." Thus, one can infer that present day knowledge of radiation chemistry will not explain everything about the radiolysis of water. Nevertheless, familiarity with the essential points of present day knowledge - and it is considerable - is germane to the problem of water decomposition in SNAP 4 reactors. Such information is important in pointing out the conditions that will minimize the problem and will show the best means for coping with it.

Under ionizing irradiation, the water molecules along the path of the incident radiation undergo dissociation. The dissociation is not uniform along the track, but rather it is concentrated in little clusters along the track. If the incident radiation is sufficiently energetic, secondary emissions, extending out like short side tracks, are also created. In the case of the heavier particle irradiation, α -particles for example, the clusters overlap creating a path approximately 20 Å wide. Allen gives an excellent pictorial description of this process.⁽¹⁾ The mechanism which explains the interaction of the incident particles and the water molecules has been studied by numerous investigators. Suffice to say for the purpose of this report that the water is dissociated into free radicals labeled H and OH. In the clusters, the free radical concentration is initially of the order of 1 molar. Naturally, at this concentration, in addition to being very reactive, many of the free radicals can react together to give H_2 , H_2O_2 or back react reform H_2O . Furthermore, some can react with the molecular produce H_2 and H_2O_2 . Finally, those that do not react quickly enough diffuse into the bulk of the fluid and react with solute molecules. It is questionable that the free radical really exist as H and OH.⁽¹⁾ At any rate, the outward evidence shows that they can always be labeled as such. It is a very interesting and important point that O radicals, or their equivalent, do not exist.

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The existence of free radical and their interaction with each other and with other molecules is inferred from the observed chemical reactions on dissolved substances in the water. Through extensive experimental investigations, it has been reasoned that the radiolysis of water can be described by the following equations.

The foregoing reactions have been studied extensively at ordinary temperatures (approximately 25°F). Reactions (1) and (3) form a rapid chain mechanism by which H_2 and H_2O_2 reform water while reactions (2), (4), (5) and (6) tend to interrupt the chain.⁽⁶⁾ Inspection of the equations shows that increasing the partial pressure of H_2 will favor reaction (1). This has been demonstrated in reactor tests where hydrogen has been added to the feedwater to reduce the net decomposition rate. Also, increasing the pressure in accordance with LeChatelier's law, will favor reaction (1) and retard reaction (6). The ratio of the rate constants for reactions (1) and (2) has been determined to be $k_1/k_2 = 1$, and that for reactions (5) and (3') to be $K_5/K_{3'} = 1.85$.⁽⁴⁾ Another reaction that is important only a higher temperature ($>200^\circ\text{C}$)⁽⁷⁾ is:

-
- (a) The hydrogen radical produced in this reaction behaves differently from the radical produced as a primary free radical and is therefore denoted as H' .^(2,3,4)
- (b) HO_2 , the hydroperoxy radical, can also be produced as a primary product in a tertiary reaction of OH radicals,^(4,5) accordingly it is denoted as a product in Equation (0).

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Through free radical reactions in aqueous solutions, the g-values (molecules per 100 e.v. of radiant energy absorbed) have been determined. The best compilation under acid conditions has been listed by Hart⁽²⁾ and is given below.

TABLE IRadiation Yields in Air-Free 0.8 N Sulfuric Acidand
Air-Saturated Water

Radiation	g(H)	g(OH)	G(H ₂)	G(H ₂ O ₂)	g(H ₂ O ₂)	Total g(H ₂ O)
<u>Air-free 0.8 N. sulfuric acid</u>						
Co ⁶⁰ γ-rays	3.65	2.95	0.45	0.80	-4.55	
Co ⁶⁰ γ-rays	3.70	2.92	0.39	0.78	-4.48	
18-mev D ⁺	2.39	1.75	0.71	1.03	-3.81	
8-mev D ⁺	1.71	1.45	1.05	1.17	-3.80	
32-Mev He ⁺⁺	1.28	1.06	1.14	1.25	-3.56	
B ¹⁰ (n, α)Li ⁷	0.23	0.41	1.66	1.57	-3.54	
<u>Air-saturated water</u>						
Co ⁶⁰ γ-rays	2.78	2.28	0.42	0.67	-3.70	-4.76
18-mev D ⁺	2.90	2.36	0.40	0.67	-3.89	-4.42
32-Mev He ⁺⁺	1.55	1.13	0.66	0.87	-2.78	-4.65
11-Mev He ⁺⁺	1.65	1.07	0.62	0.91	-2.57	-4.84

*Aerated solutions

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As shown in the table, the lighter types of radiation - γ -rays and electrons - produce greater quantities of free radicals, whereas the heavier particles produce large amounts of the molecular products. Consequently, reactions that involve free radicals are promoted by light particle irradiation. On the other hand, the heavier particles produce a greater quantity of hydrogen and hydrogen peroxide. Thus the type of irradiation can have a great effect on the quantity of water decomposed since the relative amounts of free radicals and molecular products tend to favor or disfavor certain of the chain reactions. Decomposition of water in nuclear reactors is greatly affected by this fact. In fact, if $g(H) + g(OH) > g(H_2) + g(H_2O_2)$, pure water will not decompose. The γ -ray component does much to suppress the water decomposition initiated by proton recoils. An uncertainty in the SNAP 4 design is knowing how much recombination can be credited to the γ -rays.

With G-values of the free radicals and the initial molecular products established, and the ratios of reaction rate constants of pertinent reactions, general kinetic equations⁽⁵⁾ have been set up to enable estimation of $G(H_2)$, $G(O_2)$ and $G(H_2O_2)$,^(c) the actual observed yields of the final products of decomposition. The equations are based on a mechanism of reactions (1) through (6) and are somewhat cumbersome, as would be expected with so many reactions taking place simultaneously. Since they apply to water systems at room temperature under non-boiling conditions, they cannot be applied in making an estimate of the SNAP 4 radiolytic rate.

Of particular interest to SNAP 4 design was an investigation conducted to 200°C.⁽⁷⁾ The results could be explained based on a mechanism of reactions (1), (4), (6) and (7). Reaction (7) was found to be very rapid and resulted in a reduction of the H_2O_2 concentration to less than detectable limits. The overall recombination to water could be expressed as

$$4.9 H_2 + 2.45 O_2 \xrightarrow[100 \text{ e.v.}]{\gamma} 4.9 H_2O,$$

(c) In this report, g denotes the yield of free radicals and molecular products surviving track reactions while G denotes final products.

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which in essence was equal to the production rate of primary free radicals H and OH. This illustrates the point that under proper conditions, γ -rays aid recombination.

Another article of special interest to SNAP 4 was an investigation to better understand the effect of bubbling on the decomposition rate.⁽⁸⁾ This study was prompted when the operation of BORAX III revealed a much higher decomposition rate than that observed in non-boiling reactors. In this study, CO_2 , was bubbled through water subjected to gamma irradiation. The results showed the following:

1. Molecular hydrogen was swept from the solution so effectively that no back reaction with H_2 was possible. In other words, $G(\text{H}_2) = g(\text{H}_2)$.
2. H_2O_2 decomposed yielding O_2 at a rate, $G(\text{O}_2) = 1/2g(\text{H}_2) = 1/2g(\text{H}_2\text{O}_2) = 1/4(g(\text{H}) - G(\text{OH}))$.
3. Over the range of 0.4 to 7.3, pH was found to be an unimportant variable.
4. Variation of temperature over the range of 8°C to 96°C produced little change.
5. Intensity of irradiation from 1.3×10^{20} to 1.25×10^{22} e.v./l -min had little effect.

Thus it is seen that the overriding action of the bubbles sweeping the H_2 from the system minimized the other variables which would normally be important.

Although SNAP 4 is to be operated at neutral pH, it is important to ascertain the effect of pH should the system depart from neutral conditions. After reviewing the more important studies in this area, it is difficult to assess the effect of pH per se. Varying the pH of a solution always involves the addition of extraneous ions, ions other than H^+ and OH^- . Consequently, concomitant effects are always present. Some capsule tests in the Oak Ridge X-10 reactor showed that the radiolytic rate in 10 N KOH solution was no higher than in pure water.⁽⁹⁾ A Russian investigator,⁽¹⁰⁾ using X-irradiation, presents

evidence and arguments for the ionization of OH and HO₂ radicals in alkaline solution as follows:

In addition, Hart and Platzman ⁽¹¹⁾ propose a third reaction

where e_{aq}⁻ is a hydrated electron. Intuitively, pH dependency is a result of the reactivity of the products of the foregoing three reactions, but very little is known of this reactivity. In a study involving X-irradiation of formic acid solutions (which Hart and Platzman present as evidence of the effect of pH), the variation of the g-values is not drastic over the pH range of 0.38 to 11.58. Allen summarizes his finding in a graph which shows the g-value decreasing slightly in the range of 0.5 to 3 and thereafter remaining constant to pH of 6.⁽¹⁾ He further states that adequate data are not available to extend this information into the alkaline region. By way of conclusion, it appears that pH, by itself, will not tremendously affect radiolysis in the region just greater than pH of 7; however, the data do not allow a conclusive statement to this effect.

Dissolved impurities can have a significant influence on the net decomposition rate particularly if they can act as oxidizing or reducing agents. This is due mainly to the fact that the H and OH are themselves tremendously powerful reducing and oxidizing agents. The reactions may be represented as follows:

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It is seen that this can interrupt the chain mechanism of reactions (1) and (3). A small concentration Br^- ion, as an example, will result in a decomposition rate many times that of pure water. Another example of particular importance to SNAP 4, since dissolved oxygen (from radiolysis) cannot be completely suppressed, is the reaction of dissolved oxygen and the H radical. It may be represented as follows:

whereby the reducing power of the H radical is changed over to the oxidizing power of the HO_2 radical. This reaction takes place at oxygen concentrations as small as 10^{-6} molar.⁽²⁾

This report covers a relatively small portion of the water radiation chemistry references, and they in turn represent a small portion of the total information available in the open literature. A much more thorough investigation would contribute little to the immediate problem of predicting accurately the decomposition rate in SNAP 4. Nevertheless, this limited study points up the fact that the overall decomposition mechanism is complicated, and it would be very difficult to determine the extent each variable contributes to the net rate. Thus, it is impractical to estimate the radiolytic rate from water radiation chemistry considerations alone. The literature investigation, has however been very helpful in providing an insight into the means of controlling the radiolytic rate. Briefly, they may be summarized as follows:

1. The back reactions are thermodynamically favored. If proper conditions are provided, much recombination can take place prior to the final expulsion of H_2 and O_2 from the liquid phase.
2. Increasing the total pressure of the system and increasing the partial pressure of hydrogen significantly favors the back reactions.
3. Increasing the temperature will favor the back reactions, particularly reaction (7).

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4. The bubbling action of a boiling water reactor is highly effective in sweeping the molecular gaseous products from solution. Bubbling creates a non-equilibrium condition with regard to decomposition and recombination rates, and makes the water chemistry of a boiling water reactor quite different than a pressurized water reactor.

5. Small amounts of impurities can greatly affect the decomposition rate.

6. Light particle irradiations, γ -rays for example, promote recombination.

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III. WATER DECOMPOSITION IN BOILING WATER REACTORS

The second approach to estimating the decomposition rate in SNAP 4, is the extrapolation of radiolytic rate data of other boiling water reactors to condition expected in SNAP 4. This can be done with considerable confidence. Unfortunately there are few data available on recent reactor operation. Most of the data that are reported in the literature were generated prior to the Second Geneva Conference. The data of several reports show that the net radiolytic rate is a function of total system pressure, partial pressure of hydrogen (hydrogen addition to the feedwater), temperature, specific conductivity of the water, pH, and dissolved material. Total pressure and hydrogen addition are the two most effective variables.^(12,13,14,15) Sufficient data are not available to determine clearly the independent effects of conductivity, pH, and dissolved material.⁽¹⁴⁾ Also, as mentioned in the discussion on water radiation chemistry, gamma irradiation itself aids the recombination reactions. Then too, a point seldom discussed in the literature, the fraction of energy absorbed by the water is one of the more important factors. Since the fraction of water in the SNAP 4 core is small compared to other boiling water reactors, the relative decomposition rate would be expected to be much less. The fraction of the energy absorbed in BORAX and EBWR is about 4%;⁽¹⁶⁾ the estimate for SNAP 4 is 1%.⁽¹⁷⁾

Effect of pressure is best exemplified by the data of Figure 1.⁽¹⁸⁾ The intended operating pressure for SNAP 4 is 1220 psia. Everything else being equal, its radiolytic rate would be expected to be slightly less than the rate in Vallecitos, (which operated at 1000 psia).

The next important variable which can appreciably reduce the decomposition rate is hydrogen addition. As pointed out in the previous section, it is immediately obvious that reaction (1) would be increased with increase in H_2 partial pressure. The literature is not explicit as to the actual concentration of H_2 in the core

FIGURE 1

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WATER DECOMPOSITION RATES IN EBWR (LB)

○ EBWR 600 PSIG, 448°F
□ EBWR 300 PSIG, 420°F
△ BORAX-IV 300 PSIG, 420°F
× VALLECITOS 1000 PSIG, 544°F

DECOMPOSITION RATE, LITER O₂/MIN (STP)

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solution in tests conducted in BORAX, ALPR (SL-1) and EBWR.^(12,13,16) Thus, the available data can only be used to predict the degree of suppression effected by a given amount of hydrogen by first assuming the dilution factor of the feedwater in the core. The data do show, however, that the net radiolytic rate decreases with hydrogen addition. The reduction rate decreases with increasing amounts of hydrogen. Since the concentration of the radiolytic gases in the core of SNAP 4 is estimated to be about the same as EBWR because of the higher power density of SNAP 4, there may be an appreciable reduction in the SNAP 4 net rate by adding hydrogen; however, the problems associated with collecting the hydrogen in the condenser of a boiling water reactor and re-injecting it into the feedwater are undoubtedly greater than the improvements hydrogen addition could make.^(d)

A point closely related to hydrogen addition is the fraction of hydrogen and oxygen stripped from the liquid phase by the bubbling action. EBWR tests have indicated stripping is not complete.⁽¹⁸⁾ The back reactions are favored by the γ -ray field that exists around the outside of the core, the region down through which the water passes by natural convection. For SNAP 4, circumstances will not be so favorable since all the water from the core is routed to the steam separator. It is probable that not all the dissolved gases will be expelled as the water passes through the steam separator, but the fraction expelled will certainly be higher than in EBWR. SNAP 4 conditions more closely approach Dresden in this regard since both employ force circulation. The radiolytic rate information from Dresden show that a 50% increase can be attributed to this factor.⁽¹⁹⁾

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- (d) In terms of total quantity of gas to be handled, there is little or no advantage of adding hydrogen; the reduction in radiolytic rate is offset by the hydrogen added. On the other hand, a high concentration of hydrogen has advantage in minimizing possible accumulation of explosive mixtures of hydrogen and oxygen (assuming a 100% leak-tight system).

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There are few data giving the effect of alkaline pH on the radiolytic rate. Most of the information reported applies to neutral or slightly acid water. Data from BORAX IV^(16,20) show in one case that increasing the pH from 5.7 to 8.3 significantly lowered the radiolytic rate. By contrast, in another test at constant power, the radiolytic rate decreased rapidly over the pH range of 4 to 6 after which it leveled off through to a pH of 8.3. In BORAX IV operation, the resulting pH was really a secondary effect. It was the natural consequence of removing ions from the water by the action of the demineralizer. Thus a change in the decomposition rate did not by itself indicate the radiolytic rate to be a function of pH. Surveying all the data on pH, it would appear that the net decomposition rate will be little affected by pH over the region close to neutrality.

The observed net $G(H_2)$ values have been calculated from radiolytic data of BORAX and EBWR.⁽²¹⁾ These may be termed reactor G-values to distinguish them from values for the primary products. For EBWR operating at 600 psig, $G_R(H_2) = 0.15$; for EBWR and BORAX operating at 300 psig, $G_R(H_2) = 0.21$. Information from Dresden indicated their ejector gas rate to be 23 cfm when the reactor is at full power.⁽¹⁹⁾ Assuming in-leakage of air to be negligible and the energy absorbed in the water to be 4%, the $G_R(H_2)$ for Dresden is 0.12.

The above reactor G_R values are much lower than the $G(H_2)$ values of Table I, because the back reactions are automatically taken into account. Also, these values automatically include effects peculiar to a reactor system and which may be non-existent in a laboratory test. For example, the reactor data accounts for the effect of impurities arising from corrosion products. Thus the advantage of using reactor G_R -values is immediately obvious since it circumvents the need to postulate the total effect of the many back reaction factors.

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IV. ESTIMATED SNAP 4 RADIOLYTIC RATE

As already mentioned, it is much more practical to estimate the radiolytic rate from reactor data than from water radiation chemistry data. The latter requires postulation of the recombination mechanism and estimation of such factors as fraction of the gas unstripped, concentration of H_2 , O_2 and H_2O_2 actually existing in the core solution, and rate constants of the back reactions. AI's Radiation Chemistry Department advised that the reactor data be used because of the difficulty of applying the radiation chemistry data.⁽²²⁾ However, one estimate has been included in this report based on the mechanism shown to exist when complete stripping occurs, i.e., $G(H_2) = g(H_2)$. It may be considered as the maximum possible rate. This study has shown that boiling water reactors do not approach this condition; therefore, an estimate based on complete stripping can be considered the highest possible limit.

Table II lists the estimates and the bases upon which they are made. Since the Dresden data are more applicable to SNAP 4 than the other data, the estimate based on the Dresden G_R -value is the best. The value of 17 lbs/day, based on no back reactions, sets the absolute maximum limit. Since there is certain to be some back reactions, a rate of 10 lbs/day appears as a more plausible maximum rate.

TABLE II
ESTIMATES OF SNAP 4 RADIOLYTIC RATE⁽¹⁾

Basis	G-Values	Back Reaction Mechanism	Water Decomposed lbs/day	Rating
BORAX, EBWR & VBWR gas rates corrected to SNAP 4 conditions of pressure and energy absorbed	Not required for estimate	Accounted for as occurs in natural circulation reactors	2.5	Probably slightly low since force circulation not taken into account
GR ⁽²⁾ of BORAX & EBWR corrected to SNAP 4 conditions of pressure	GR(H ₂) = 0.09 (at SNAP 4 conditions)	Accounted for as occurs in natural circulation reactors	3.8	Probably slightly low since force circulation not taken into account
Dresden gas rate with a correction factor for SNAP 4 pressure	Not required for estimate	Accounted for as occurs in a force circulation reactor	4.2	Next best estimate
GR value for Dresden corrected to SNAP 4 pressure	GR(H ₂) = 0.12 (at SNAP 4 conditions)	Accounted for as occurs in a force circulation reactor	4.7	Best estimate (suggest adding contingency factor of 100%)
Theoretical yield of H ₂ as a primary product at about the middle of the energy spectrum	GR(H ₂) = 1.0	Complete stripping of H ₂ and O ₂ assumed, ² GR(H ₂) = g(H ₂)	43	Possible but highly improbable
Estimated GR for no recombination	GR(-H ₂ O) = 0.4	None, no credit taken for γ-ray recombination	17	Possible

- (1) All estimates have been made on the basis of 1% of the energy being absorbed in the water and reactor power at 12 Mw(th).
- (2) GR is notation for "reactor G-value", or the molecules of water decomposed per 100 e.v. of energy absorbed in the water of a reactor as actually observed or estimated.

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V. HANDLING OF THE RADIOLYTIC GAS

In the foregoing section the decomposition rate was established as most probably of the order of 5 lbs/day. As the most plausible maximum rate, a value of 10 lbs/day appears to be realistic. There are really only two basic ways of handling the decomposition gases. The method employed by all boiling water reactors to date is discharging these gases to the atmosphere. Prior to discharge, the gases are diluted to avoid accumulating an explosive mixture. In addition, they are monitored to insure that radiological disposal standards are met. If the reactor system is not hermetically sealed, and there is leakage from the turbine or other parts of the system, there may be merit in discharging directly to the atmosphere. For a hermetically sealed system, this approach is completely unacceptable. Since one of the long range goals is to make SNAP 4 a hermetically sealed system, a recombiner system is the only realistic way open to SNAP 4.

A. BASIS FOR DESIGN OF THE SNAP 4 RECOMBINER

Design of fixed bed catalytic reactors requires the application of mass transfer phenomena coupled with heterogeneous chemical kinetics. In fixed bed catalytic systems, four types of sequential steps take place.

1. Mass transfer of reactants and products to and from the gross exterior of the catalyst particle surface and the main body of the fluid.
2. Diffusion of reactants and products into and out of the pore structure of the catalyst when the reaction takes place at interior surfaces.
3. Adsorption of reactants and desorption of products on active sites of the catalyst surface.
4. The surface reaction of adsorbed reactants to produce adsorbed products.

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Generally one of these steps is so much slower than the others that it becomes the "pace-setter" or rate controlling mechanism. Obviously, when more than one of the steps are equally slow, the problem becomes immensely more complicated. Also, each step is a function of many design and operating variables, and it is possible that the rate controlling mechanism can shift from one step to another simply by changing a single variable, temperature for example. To develop equations for prediction of catalyst bed volume or performance it is necessary, (1) to determine or postulate the rate controlling mechanism, (2) develop rate equations which properly express that mechanism, and (3) apply the rate expressions to a mass balance representing the steady state flow situation under study. Generally, when catalyst bed equations are developed, they are quite impractical because they involve terms to which it is difficult or impossible to assign numerical values. Frequently, these terms are reaction rate constants, chemical equilibrium constants, catalyst surface area, and catalyst activity.

With regard to the design of the SNAP 4 recombiner, there are available the data of recombiners of other nuclear reactors. One such thorough and practical treatment is that of H. J. Garber and F. N. Peebles (23) who have endeavored to develop equations for catalyst volume in terms of easily obtainable parameters. This treatment presents equations for five rate determining mechanisms and then goes on to show that the HRE recombiner development data of J. A. Ransohoff and I. Spiewak (24) correlated best by use of a "Dual-Langmuir mechanism". The "Dual Langmuir mechanism" postulates that oxygen diffusion is rate-controlling in the first portion of the bed, and hydrogen diffusion is controlling in the last portion of the bed, and hydrogen diffusion is controlling in the last portion of the bed, with the change-over occurring at the point when oxygen and hydrogen are diffusing to the catalyst surface in their stoichiometric ratio.

The mechanism also postulates that both hydrogen and oxygen are adsorbed on the platinum; however, in the region where oxygen diffusion is controlling, recombination takes place only on those sites occupied by oxygen and which are subsequently hit by hydrogen molecules. The reverse process is assumed to take place in the latter portion of the bed where hydrogen diffusion is controlling.

With regard to the SNAP 4 recombiner, the problem is to select the mechanism that is likely to exist. Two different conditions are envisioned as possible. One could be the existence of a hydrogen rich system. This could be the result of the corrosion process using up oxygen leaving an excess of hydrogen. The other would be the existence of the gases in their stoichiometric ratio.^(e) For the hydrogen rich system, oxygen diffusion would be rate controlling since it is the constituent in smaller quantity. In the latter case, oxygen diffusion would be rate controlling since it is the constituent in smaller quantity. Consequently, for both cases oxygen diffusion would be expected to be rate controlling throughout the entire length of the bed. For oxygen diffusion controlling, the mechanism becomes the "Langmuir mechanism" as termed by Garber and Peebles. The equation presented by these authors for predicting catalyst volume for conditions under which the "Langmuir Mechanism" applies as follows:

$$aL = \frac{V_o}{K_1 J} \left[2H_o f_2 - (2O_o - H_o) \right] \ln \frac{1}{1-C} \quad (1)$$

- - - - -

- (e) Until a stabilized oxide film is formed, the corrosion process will take up oxygen, causing the gas composition to depart from the stoichiometric ratio. The case here assumes that the up-take of oxygen has been completed in prior reactor operation and that the excess hydrogen has been drawn from the system.

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where a = cross-sectional area of the bed, ft^2
 L = length of the bed, ft
 V_o = total fuel rate entering the bed, lb-moles/sec
 H_o = mole fraction of hydrogen in the feed stream
 f_2 = fraction of hydrogen reacted
 O_o = mole fraction of oxygen in the feed stream
 C = fraction of oxygen converted to water
 J = catalyst surface area, ft^2/ft^3
 K_1 = mass transfer coefficient, $\frac{\text{lb moles}(\text{hydrogen mole fraction})}{(\text{sec})(\text{ft}^2)(\text{oxygen mole fraction})}$

All the values that go into the above equation are either given by the conditions of the problem, or can be determined rather easily. The most difficult function to determine is K_1 , the mass transfer coefficient. Here, Garber and Peebles use the method of Gamson⁽²⁵⁾ which in essence is a dimensionless group correlation involving the Schmidt and Reynolds numbers.

There are arguments for and against the use of Garber and Peebles equation for predicting SNAP 4 recombiner size. A major factor in favor is the similarity of the proposed recombiner system with the one upon which the Garber and Peebles treatment is based. The catalyst, platinized alumina pellets, proposed for SNAP 4 is identical to that used in the HRE recombiner development program. Also, both are hydrogen-oxygen-water systems. In addition, the fact that the treatment is based upon some experimental data makes it far superior to a treatment which is based only on theoretical considerations. A weakness of the Garber and Peebles treatment with regard to SNAP 4 application is its failure to take into account the diluting effect of steam. Inspection of the equation will show that increasing the percentage of steam in the fluid (smaller values of H_o and O_o) will actually result in smaller catalyst volume. Intuitively this is not

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right. Decreasing reactant concentration axiomatically leads to a reduction in reaction rate, and a reduced reaction rate normally requires a larger catalyst volume.

B. CALCULATION OF THE SNAP 4 RECOMBINER VOLUME

With full recognition of the weaknesses of the Garber-Peebles treatment with regard to its application to SNAP 4 recombiner design, it remains the best available approach to us at present. Calculations were performed employing the equation under several different assumed conditions for SNAP 4. These calculations endeavored to find if the Garber-Peebles equation was sensitive to any particular operating variable. The results, which are shown in Table III, did not point up a particular sensitivity but, as mentioned earlier, did show a failure to take into account the diluting factor of the steam. All cases were calculated on the basis of oxygen conversion of 98%.

C. DISCUSSION OF THE RESULTS

The bed lengths for the cases involving a stoichiometric ratio of gases (no excess hydrogen) are so much smaller than the others, an explanation is needed. The postulated mechanism allows a possible explanation. The mechanism postulates that oxygen diffusion to the catalyst surface is rate controlling and also that the hydrogen absorption offers a retarding effect. In essence, the mechanism requires that the oxygen molecules must diffuse to the catalyst surface and find unoccupied sites (sites not already occupied by hydrogen). Then the oxygen must subsequently be hit by a hydrogen molecule. The water molecules that are formed by this process then leave the surface of the catalyst. The water molecules (steam) of themselves do not offer any restraint upon the mechanism. Accordingly, the catalyst volume requirements are less for the stoichiometric gas ratio than for an excess of hydrogen.

TABLE III
CALCULATED CATALYST BED VOLUME UNDER VARIOUS CONDITIONS
(Oxygen Conversion 98%)

Excess Hydrogen cc/kg Steam	Decomp.Rate lbs/day	Total Feed Rate lbs/day	Hydrogen in Feed Vol.%	Steam in Feed Vol.%	Recomb. Pressure psia	Avg.Cat. Bed Temp. °F	Catalyst Bed Dimensions Dia.(in.) Lgth.(in.)	
30	3	133	16.8	82	23	450	3	5.7
60	3	136	27.6	72	23	450	3	10.2
30	3	264	16.8	90	23	450	3	4.0
30	6	157	16.1	82	23	450	3	7.3
30	3	147	1.76	98	90	450	3	1.7
30	3	133	16.8	82	23	700	3	0.9
0	5	147	3.33	95	23	450	3	0.7
0	5	147	3.33	95	23	450	1	1.5
0	10	152	6.34	91.5	23	450	3	0.9

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The proposed recombiner size is 3 in. diameter by 12 in. long. This covers the largest calculated dimensions (the case for excess hydrogen of 60 cc/kg of steam) and many times larger than that required for the more likely conditions, or stoichiometric conditions. It is believed that selecting the aforementioned size adequately covers the many contingencies that exist. Since the unit is small and relatively inexpensive, there exist no stringent reasons for designing close to the minimum required volume. Furthermore, it is to be noted that raising the temperature greatly reduces the needed catalyst volume. It is suggested that increasing the temperature be the principal operating control to increase recombination capacity should it become necessary. There is one further point worthy of mentioning. The experimental data upon which the Garber-Peebles treatment is based are characterized by an inlet steam composition of 87-93%, the same magnitude as anticipated for the SNAP 4 recombiner.

D. RECYCLING AND GAS BUILD-UP

Since the recombiner design is based upon a conversion efficiency of 98% of the hydrogen and oxygen entering the recombiner, the question arises as to what are the consequences of uncombined 2% of the gases upon a recycling system. Could these gases build up to an undesirable high concentration? Particularly, this situation might develop if the performance of the recombiner should decrease with increasing load. In answer to this question, a detailed analysis of the several factors that affect recombiner performance was undertaken.

Consider again the Garber-Peebles equation for the Langmuir mechanism with oxygen diffusion rate controlling. For convenience the equation is recopied:

$$aL = \frac{V_o}{K_1 J} \left[2 H_o f_2 - (2O_o - H_o) \right] \ln \frac{1}{1-c} \quad (1)$$

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Since it is anticipated that SNAP 4 will not employ excess hydrogen to suppress radiolytic decomposition, the hydrogen and oxygen entering the recombiner will be essentially at their stoichiometric ratio.^(f) Under these conditions, the " $2O_2 - H_2$ " term of Equation (1) becomes zero, significantly simplifying the above equation. The quantity of hydrogen recombined, $V_{O_2} f_2$, becomes:

$$V_{O_2} f_2 = 1/2aLJK_1 \quad (2)$$

However, for any given recombiner, the expression $1/2aLJ$ is a constant so that the quantity of hydrogen recombined is directly proportional to K_1 . Consequently, the change in capacity of the recombiner can be determined by learning how the mass transfer coefficient is affected by changes in operating variables. It should be mentioned at this point that recombination efficiency as might be noted by f_2 , the fraction of hydrogen undergoing recombination, cannot be properly evaluated per se. For cases of catalyst volume many times larger than the required volume, as has been proposed to cover contingencies and as discussed in the previous section, the solution of Equation (2) leads to the f_2 greater than 1.0, an absurd condition. Therefore, change in recombiner performance can best be evaluated by studying changes in capacity rather than the fraction of hydrogen converted.

All the values of the foregoing except K_1 are given by the conditions of the problem and the choice of catalyst. Regarding the mass transfer coefficient, K_1 , Garber and Peebles found it was best expressed by the following relationship:

$$K_1 = \frac{2.457 \times 10^{-4} G (1-v)^{0.2} T_g e^{-933/T_g}}{M_m Sc^{2/3} Re^{0.41}} \quad (3)$$

where the newly introduced terms are:

(f) Until a stabilized oxide film is formed, the corrosion process will take up oxygen, causing the gas composition to depart from the stoichiometric ratio. The case here assumes that the up-take of oxygen has been completed in prior reactor operation and that the excess hydrogen has been drawn from the system.

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G = superficial mass velocity, $\text{lbs/ft}^2 - \text{sec}$

v = void fraction of the catalyst bed

T_g = mean gas stream temperature, $^{\circ}\text{K}$

M_m = mean molecular weight of the gas stream

Sc = Schmidt number

Re = Reynolds number

Consequently, since recombiner capacity is proportional to K_1 , changes in capacity can best be analyzed by determining the effect upon K_1 by all the factors on the right of Equation (3). This has been done by treating K_1 as an exact differential as

$$dK_1 = \left(\frac{\partial K_1}{\partial G}\right) dG + \left(\frac{\partial K_1}{\partial M_m}\right) dM_m + \left(\frac{\partial K_1}{\partial Sc}\right) dSc + \left(\frac{\partial K_1}{\partial Re}\right) dRe \quad (4) \quad (g)$$

The partials of Equation (4) were obtained by partial differentiation of Equation (3) with respect to each particular variable. For example,

$$\left(\frac{\partial K_1}{\partial T_g}\right)_{G, M_m, Sc, Re} = \frac{2.457 \times 10^{-7} (0.907) G}{M_m Sc^{2/3} Re^{0.41}} e^{-933/T} \left(\frac{933}{T_g} + 1\right) \quad (5)$$

The differentials, dG , dT_g , dM_m , etc., were evaluated from expressions that define these particular variables in terms of the problem. To illustrate, T_g , the temperature of the gas stream is a function of the quantity of gas being recombined. Considering the heat of reaction for $H_2 + 1/2 O_2 \rightarrow H_2O$ to be 58,000 cal/g-mole, the specific heat of the resulting steam to be 0.47 BTU/lb, the weight rate to be 5 lbs/day, of radiolytic hydrogen and oxygen, and the diluting steam to be 142 lbs/day, then

$$dT_g = \frac{58,000 (1.8) dW}{(0.47) (147)(18)} = 84.0 dW ^{\circ}\text{F} \text{ or } 46.5 dW ^{\circ}\text{K} \quad (6)$$

(g) For simplicity in writing, the subscripts of each partial are omitted.

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where dW is the differential change in weight rate. (The factor of 1.8 converts from cal/g-mole to BTU/lb-mole while 18 converts from lb-moles to lbs.) Likewise the other differentials were also derived in terms of dW . For Sc and Re , the procedure was quite involved. First, an exact differential equation was set up for each of these terms similar in form to Equation 4, and then the differentials were solved in terms of dW . To keep the process from becoming too formidable, secondary interaction terms were dropped.

When the arithmetic values for the partials are inserted in Equation (4) the resulting equation is:

$$dK_1 = 2.07 \times 10^{-4} dG + 4.02 \times 10^{-8} dTg - 4.03 \times 10^{-7} dMm - 6.70 \times 10^{-6} dSc - 3.59 \times 10^{-8} dRe \quad (7)$$

The values for the five differentials in terms of dW are as follows:

$$dG = 2.36 \times 10^{-4} dW$$

$$dTg = 46.5 dW$$

$$dMm = -0.0292 dW$$

$$dSc = -0.023 dW$$

$$dRe = -6.79 dW, \text{ where } W \text{ is water radiolysis weight rate, } 5 \text{ lbs/day.}$$

When the above differentials are substituted into Equation (7), the resulting expression of dK_1 is

$$dK_1 = +2.3 \times 10^{-6} dW$$

It is to be noted that the three negative differentials also have negative coefficients so that all five terms operate positively on K_1 . For the recombiner proposed for SNAP 4, namely: 3 inches in diameter, a loading of 5 lbs/day of stoichiometric hydrogen and oxygen, 142 lbs/day of diluting steam, and operating at 450°F minimum; K_1 is calculated to be 0.750×10^{-5} . Therefore, for a 1% increase in W , the corresponding increase in K_1 would be 1.5% indicating there would be

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no detrimental decrease in performance of the recombiner. Considering the conversion to remain at 98% (a conservative estimate in view of the foregoing analysis), a simple material balance of the recycle circuit shows that the steady state load to the recombiner would only be increased by 2%.

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VI. OTHER CONSIDERATIONS

There are a few other considerations relating to the operation of a recombiner in a nuclear reactor system which deserve mentioning. These are lesser problems which could nevertheless be the cause of failure of the system if not properly handled.

A. EXPLOSION HAZARD

The radiolytic hydrogen and oxygen can, under certain circumstances, result in an explosive mixture. In addition, recombiners can serve as an ignition point since the exothermic reaction can result in a very hot catalyst surface. The way of eliminating such a hazard is to prevent the accumulation of an explosive mixture of gases. For SNAP 4, this can easily be accomplished by diluting the mixture with sufficient steam. With 90% dilution of steam, the intended mode of operating the recombiner, the recombiner feed stream is well within the non-explosive region. The concern is assurance that such conditions will always prevail.

There are two events that could result in an explosive mixture being formed. One is excessive condensation of the steam, thus removing the diluent from the mixture. A sudden decrease in the turbine condenser temperature could produce such a situation; however, since it is usual to operate the turbine at the lowest temperature attainable under the particular operating conditions, a further drop in temperature is unlikely. Nevertheless, if there is any possibility that such an event might occur, proper dilution can be maintained by employing a mass flow rate controller which would hold the steam rate above a predetermined rate. The second event that could result in an explosive mixture is the failure of the recombiner to react the oxygen and hydrogen. Since SNAP 4 will be an all stainless steel system with high purity water as the working fluid, there will be no materials present to permanently poison the catalyst. The only conceivable thing

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that could impair the catalyst is the possibility of it becoming wet. Maintaining the catalyst in a dry condition requires only that the temperature is above the saturation temperature. Installing a small heater in the catalyst bed and providing the necessary instrumentation to insure that the bed is above the saturation temperature whenever the reactor is in operation can preclude the wetting problem. Thus the two events that could result in an explosive condition are easily dealt with.

B. ACCUMULATION OF NON RECOMBINABLE GASES

One of the best evidences that non-recombinable gases do not accumulate in a hermetically sealed nuclear reactor system is experience itself. Atomics International has designed and built six L-54 aqueous homogeneous reactors. These are completely sealed systems employing the same type platinum-coated alumina catalyst as proposed for SNAP 4. Following the "shake-down" operation of the first L-54, the catalytic systems have performed very successfully in all six reactors. None have experienced pressure build-up within the system during long-term operation.

The recombiner reacts the hydrogen and oxygen in an amount equivalent to the constituent in lowest stoichiometric quantity. In other words, the recombiner cannot adequately handle the non-condensable gases when they occur in a non-stoichiometric ratio, or when gases other than hydrogen and oxygen are present. Even so, the recombining system would continue to work satisfactorily if the ratio does not depart from stoichiometric excessively.

1. Side Reactions in the Radiolytic Decomposition Process

With high purity water, the net decomposition reaction yields stoichiometric hydrogen and oxygen.⁽²⁶⁾ It is a virtual certainty that these conditions will occur since the water in the reactor system will be of the highest quality and will be contained in an immaculately

clean stainless steel system. the possibility of side reactions producing a non-stoichiometric ratio of hydrogen and oxygen are so minimal that the problem can be safely ignored.

2. Incomplete Removal of Air From the System on Initial Startup

If all the air is not completely removed from the system on startup, stoichiometric conditions will be violated at the very beginning. This situation can be overcome by thorough purging with steam. In addition, all water added to the system can be thoroughly degassed. Consequently, this problem can be overcome simply by prescribing the proper checkout and startup procedures

3. Oxygen Take-Up in the Corrosion Process

The corrosion process, being an oxidation process, will consume oxygen if the only source of oxygen is radiolysis, an excess of hydrogen will result. The principal way of overcoming this difficulty is to establish a stabilized oxide film on the stainless steel surfaces before initiation of regular operation. This could be done during factory checkout operation prior to delivery of the plant. Moreover, the system can tolerate a considerable excess of hydrogen. If the condenser operating pressure is 25 psia, the hydrogen concentration can build up to 11 cc/kg of condensate and remain dissolved in the feedwater. In fact, some buildup in hydrogen would be beneficial since it would aid in suppressing the radiolytic rate. However, allowing it to build up to the point where it overburdens the non-condensable exhauster is to be avoided.

4. Diffusion of Hydrogen From the Hydrided Fuel

With a build-up of hydrogen to a concentration of 11 cc (STP)/kg of condensate, the partial pressure of hydrogen in the core coolant would be approximately 0.12 psia. The dissociation pressure, the hydrogen in the hydrided fuel is estimated to be 0.0037 psia. Thus there is little likelihood of increased hydrogen from this source.

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C. ALTERNATIVE TO HERMETIC SEALING

If the reactor system is not hermetically sealed - if there is leakage from the turbine or other points in the system - the need to recombine the radiolytic gases is greatly relaxed. Should the leakage rate far exceed the decomposition rate, there may be merit in venting the radiolytic gases along with all the other non-condensables rather than attempting to recombine and recover the resulting water. Whether this approach is taken depends upon exactly what the leak rate is. Nevertheless, since Atomics International's long term goal is a hermetically sealed system, inclusion of a recombiner on all systems will gain us considerable experience.

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