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REACTION OF GASEOUS IMPURITIES
IN A HIGH TEMPERATURE GAS-COOLED REACTOR

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ABSTRACT

The Graphite Oxidation Program (GOP) computer code has been updated to study the gas - graphite reactions in an HTGR. The effect of radiolytic as well as thermal reactions on the steady-state gas compositions of an operating HTGR is determined. The code is also useful in calculating the possible consequences of various reactor accidents. Water injection tests at the Dragon reactor have been analyzed with the code.

TABLE OF CONTENTS

LIST OF FIGURES	iv
LIST OF TABLES	iv
INTRODUCTION	1
POTENTIAL CHEMICAL REACTIONS	2
Thermal Chemistry	3
C + H ₂ O Reaction	3
C + CO ₂ Reaction	4
CO + H ₂ O $\rightleftharpoons$ CO ₂ + H ₂	4
Diffusion Effects	5
Radiation Chemistry.	6
MATHEMATICAL ANALYSIS	9
Geometry	9
Computer Calculation	11
Steady-State Calculation	13
RESULTS	14
Dragon Injection Experiment	14
Steady-State Analyses in Fort St. Vrain	15
Thermal Reactions Only	15
Effect of Radiolytic Reactions.	19
Variation in Water Inleakage Rate.	19
Accident Analysis in Fort St. Vrain	21
CONCLUSIONS.	21
ACKNOWLEDGMENT.	23
REFERENCES	24

LIST OF FIGURES

1. Efficiency of Energy Transfer	8
2. Fuel Element	10
3. Effective Area - Graphite to Coolant Hole	12
4. Dragon Reactor - Water Injection Test - August 1966	16
5. Dragon Reactor - Water Injection Test - August 1966	17
6. Steady-State Water Level vs. Leak Rate	20
7. HTGR Accident Simulation Analysis	22

LIST OF TABLES

I. G-Values for Reactions	7
II. Description of Four Zones in HTGR	11
III. Steady-State Gas Compositions in HTGR Thermal Reactions Only	18
IV. Steady-State Gas Compositions in HTGR Thermal and Radiation Reactions	19

REACTION OF GASEOUS IMPURITIES
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INTRODUCTION

A high temperature gas-cooled reactor (HTGR) is a steam generating, helium cooled, graphite moderated, power reactor. Small quantities of impurities are inevitably present in the helium coolant from inleakage of H_2O from the steam generator and from outgassing of the graphite core. At temperatures approaching $1000^\circ C$ as in an HTGR, these impurities react relatively rapidly with the graphite core and other reactor components. The reaction of water vapor with the graphite causes a buildup of CO and H_2 directly and a buildup of CO_2 indirectly by homogeneous reactions involving CO and H_2O and homogeneous or heterogeneous decomposition of CO.

Graphite corrosion and carbon decomposition must be controlled for the safe and economical operation of an HTGR. To realistically establish limits for water inleakage it is necessary to understand the rates of the possible reactions and the steady-state impurity concentrations that would be obtained. Furthermore, safety considerations require information on the possible consequences of accidental heat exchanger rupture or loss of coolant flow.

The purpose of this study is to determine steady-state gas concentrations in an HTGR. Because of the many complicating features of the reactor, intuitive estimates or rough approximations often lead to erroneous conclusions. A detailed analysis is required to avoid these errors.

This type of analysis can be accomplished by digital computation in which the code treats several possible chemical reactions, and simulates reactor temperature profiles, diffusion coefficients, radiation effects, and energy transfer from the radiation-excited helium coolant. Although the present treatment is far from optimum, attempts have been made to be as accurate and complete as possible with the fundamental data currently available. In some cases where the data are not available, estimates are made to determine the importance of the effect upon the conclusions.

This paper presents the data obtained when the GOP (Graphite Oxidation Program) computer code is used to analyze the results of the water injection experiments at Dragon reactor.⁽¹⁾ In addition, predictions of steady-state gas compositions and some accident analyses are made in a reactor with conditions similar to the Fort St. Vrain reactor.

POTENTIAL CHEMICAL REACTIONS

In a high temperature gas-cooled reactor, water leaks from the heat exchanger are the primary source of impurities in the gas system. The reaction of water vapor with the graphite will result in the formation of CO and H₂ and secondary reactions result in still other species. The following gaseous species are considered: H₂O, H₂, CO₂, and He. These gases, in contact with graphite may be involved in the following reactions:

Reactions (1) through (4) are the only ones treated in the present analysis.

Each of the reactions may be initiated radiolytically as well as thermally and rate expressions for both types of initiation are considered in the computer analysis.

THERMAL CHEMISTRY

Equations (1) through (4) show only the stoichiometric changes involved and are not intended to yield information about rate expressions or mechanisms. The analysis requires a knowledge of the rate expressions for each reaction and, in the case of the gas - solid reactions, diffusion effects must also be treated. The details of the mechanisms of these reactions and rate expressions are described in the following paragraphs:

(1) C + H₂O Reaction

The rate expression for the water - graphite reaction as determined by Giberson and Walker⁽²⁾ is given by the equation below:

$$\frac{d(-C)}{dt} = \frac{k_1 P_{\text{H}_2\text{O}}}{1 + k_2 P_{\text{H}_2}^{1/2} + k_3 P_{\text{H}_2\text{O}}} \quad (8)$$

This rate expression is consistent with the following mechanism if stationary state conditions are assumed:

where the C_f represents a free-site on the carbon surface and $C(O)$ and $C(H)$ represent adsorbed species.

(2) C + CO₂ Reaction

While there is disagreement in the literature on the appropriate mechanism for the $C + CO_2$ reaction,^(3,4) the rate expression derived from either mechanism and determined experimentally is the following:

$$\frac{d(-C)}{dt} = \frac{k_1 P_{CO_2}}{1 + k_2 P_{CO} + k_3 P_{CO_2}} \quad (12)$$

Reactions (3) and (4) considered together constitute the water - gas shift equilibrium process. The kinetics and mechanism of these reactions have been studied by Graven and Long⁽⁵⁾ and by Tingey.⁽⁶⁾ For the forward reaction the rate expression is:

$$\frac{d(-CO)}{dt} = \frac{d(-H_2O)}{dt} = \frac{k P_{CO}^{1/2} P_{H_2O}}{(1 + a P_{H_2O})^{1/2}} \quad (13)$$

where a and k are constants.

For the reverse reaction [Equation (4)] the rate expression is:

$$-\frac{d[\text{CO}_2]}{dt} = k_1 P_{\text{CO}_2} P_{\text{H}_2}^{1/2} + k_2 P_{\text{CO}_2} P_{\text{H}_2}^{1/3} \quad (14)$$

Diffusion Effects

In the case of the heterogeneous reactions [Equations (1) and (2)] it is necessary to consider the effect of in-pore diffusion. The rate expressions [Equations (8) and (12)] are valid only for uniform reaction within the graphite. At a temperature where the reaction rate is limited by diffusion, a correction term must be applied.

The diffusion limited rate is given by:

$$R_{dl} = \eta R_h \quad (15)$$

where R_h = the homogeneous reaction rate [Equation (8) or (12)] and η is the diffusion correction and has a value ≤ 1.00 . The value of η depends on the effective diffusion coefficient, the graphite temperature, the geometry of the system, the total gas pressure, and to a lesser extent the gas composition. The effective diffusion coefficient, Z , is defined:

$$Z = m D_{12} \quad (16)$$

where D_{12} is the mutual gaseous diffusion coefficient for $\text{H}_2\text{O}/\text{He}$ or CO_2/He and is a function of total pressure and temperature; "m" is a constant for a particular graphite and has been related to the tortuosity and porosity of the graphite. Values of m ranging from 0.01 to 0.0004 have been reported.^(7,8) The particular value chosen has a significant effect on the results.

RADIATION CHEMISTRY

Any comprehensive evaluation of reactions in a reactor system must treat radiation-induced reactions as well as thermally-induced ones. Often the rates of the radiation-induced reactions equal or even exceed the rates of the thermal processes. Although the radiation-induced processes are not nearly as well understood as the thermal ones, some data are available on the rates of attack by excited CO_2 and H_2O on graphite⁽⁹⁾ as well as on the homogeneous gas phase reactions of CO_2 with H_2 ⁽¹⁰⁾ and CO with $\text{H}_2\text{O}(\text{g})$.⁽¹¹⁾

Table I lists the 100 eV yields ($\underline{G}$ values) of the various reactions of interest. All of these data are determined in a gas mixture containing only the gaseous reactants and in some cases an inert gas. A complicating feature in accurately treating the radiation-induced reactions, is the lack of information on energy transfer from excited helium to the reactive impurity. Under normal HTGR operating conditions, essentially all of the energy dissipated is absorbed initially in the helium, and then may be transferred to the reactive gas. The efficiency of this energy transfer at very low impurity content depends upon the impurity concentration. In the multi-component reactor system, one must also be concerned with the manner in which the energy is partitioned between the gas components and the effectiveness of the energy in producing the various possible products. Since information is not available on the partitioning of the energy, it was divided among the components according to their relative partial pressures.

TABLE I
G-VALUES FOR REACTIONS

<u>Reaction</u>	<u>G (Molecules/100 eV)</u>
C + H ₂ O	0.35
C + CO ₂	1.35
CO + H ₂ O	$79.7e^{-2,500/RT}$
CO ₂ + H ₂	$13.7e^{-1,620/RT}$

Data from the Dragon Project⁽⁹⁾ indicate that for the CO₂- graphite reaction about 50% of the energy is transferred when the CO₂ level is 10 to 50 ppm.

In the absence of more quantitative information, we have expressed the efficiency of the energy transfer from excited helium to impurity gases as follows:

$$E = 1 - e^{-r\sum P_i}$$

where r is a constant used to adjust the function to the desired level and P_i , the impurity partial pressures, are summed over all impurity gases. For the purpose of this work, r has been chosen as one and the P_i are expressed in units of millimeters of mercury. A plot of this function for impurity concentrations between 0 and 100 ppm is shown in Figure 1.

The activated molecule is then partitioned between the possible reactions by the expression $g_j/\sum g_j$ where g_j denotes the G(value) of the various radiolytic reactions involving the species.

The overall rate of the radiation-induced processes are thus expressed as follows:

FIGURE 1
Efficiency of Energy Transfer

$$(dp_i/dt)_j = g_j \delta E_{ij} \quad (17)$$

$(dp_i/dt)_j$ = change in partial pressure of the i th component
by reaction j in units of molecules/cm³-second

g_j = molecules reacted or formed per 100 eV of energy
absorbed, for a specified radiolytic activated
reaction

δ = radiation dose rate, 100 eV/cm³-second

E_{ij} = efficiency factor to account for energy transfer
from helium to species i and alternate paths j
of reaction for that species

MATHEMATICAL ANALYSIS

The Graphite Oxidation Program is capable of handling the geometry of the HTGR, including recirculation of the gas through four different zones of temperatures and gas flow rates.

GEOMETRY

The HTGR core* consists of a number of hexagonal bricks containing coolant and fuel holes. The arrangement of a typical brick is shown in Figure 2. A cluster of seven bricks makes a region (six of the outermost regions contain only five bricks). There are thirty-seven regions in the core. A number of regions are grouped together to form zones. Each zone has a unique gas flow rate and graphite temperature distribution. During

*This information is for the Fort St. Vrain reactor. The Dragon reactor is of similar, but smaller, design.

the six-year life of the core, each region will be subjected to all of the zones of temperature and flow. Table II describes the four zones.

TABLE II
DESCRIPTION OF FOUR ZONES IN HTGR

Zone No.	I	II	III	IV
Flow Rate (lb/sec)	0.061	0.065	0.035	0.020
Fraction of Total Core	0.006	0.167	0.680	0.148
Coolant - Graphite Interface Temperatures	°C	°C	X/L*	°C
	403	403	0.00	403
	403	403	0.07	403
	527	499	0.14	487
	552	514	0.20	504
	693	622	0.31	607
	824	720	0.43	703
	900	775	0.54	762
	960	821	0.66	810
	994	848	0.77	839
	1046	887	0.83	877
	931	799	0.92	799
	931	799	1.00	799

*X/L = fractional distance down coolant channel.

The dimensions of the gas flow channel and the effective amount of graphite per coolant channel must be known for the analysis. The latter is calculated on the basis of the relative areas of coolant holes to total graphite cross section. For GOP the number is expressed as the length of the side of a square whose area is equivalent to the cross sectional area of a hollow cylinder sans holes, see Figure 3.

COMPUTER CALCULATION

Input to the computer consists of geometry factors, graphite temperatures, gas flows, and chemical reaction rate constants.

FIGURE 3
Effective Area - Graphite to Coolant Hole

A typical calculation consists of specifying the initial partial pressures of H_2O , CO , CO_2 , and H_2 . The change in the partial pressures are calculated along the length of the coolant channel for each of the four zones. The partial pressures at the exit are averaged and then corrected for water leak rate and gas purification and the calculation repeated. The number of cycles to be run is specified as input. In addition to calculating the gas compositions at any point in the reactor, a profile of the graphite oxidation is also obtained.

A complete program listing is available from the authors on request.

STEADY-STATE CALCULATION

Although it is possible to calculate the transitory conditions during reactor startup, shut down, or changes in leak rate and purification rate, it is not practical to do so for extended time periods because of the computer time required. Therefore, the steady-state water concentration is determined from the assumption that at steady-state the water inleakage rate is equal to the rate of water removal by all processes, i.e.,

$$\text{Leak Rate} = \text{purification rate} + \frac{d[-H_2O]_{ss}}{dt} \quad (18)$$

where $\frac{d[-H_2O]_{ss}}{dt}$ represents the change in water level, at steady-state, as a consequence of the various chemical reactions. Under normal operating conditions, the leak rate and purification rate are constants. The term $\frac{d[-H_2O]_{ss}}{dt}$ is estimated by choosing conditions somewhat different than steady-state. The GOP program is then used to calculate a value for $\frac{d[-H_2O]}{dt}$ for the chosen conditions. This rate which is a function of H_2O concentration is then used to re-estimate the steady-state water

concentration by equation (18). The calculation is repeated until the changes in concentration reach a small predetermined value.

A similar approach is taken to evaluate the steady-state concentrations of H_2 , CO_2 , and CO . However, these species occur only due to chemical reaction, since no inleakage is assumed.* They are removed by the purification as well as by chemical processes.

RESULTS

The present analysis was performed to determine three things:

1. The steady-state gas composition expected as a result of the first water injection test in Dragon.
2. The steady-state H_2/H_2O ratios for normal operation of the Fort St. Vrain reactor.
3. The possible pressure buildup in the Fort St. Vrain reactor as a consequence of a major heat exchanger leak.

DRAGON INJECTION EXPERIMENT

The Dragon reactor is very similar in design to the Fort St. Vrain, though on a smaller scale. Dragon can be divided into four concentric zones which all have the same flow rate. The two centermost zones also have identical temperature profiles.

In August 1966, water vapor was deliberately added to the helium gas stream of the Dragon reactor. The water vapor was added first as a large

*The program permits the inclusions of a hydrogen inleakage rate for a special application. The value for normal analysis would be set equal to zero.

surge ($\sim 10 \text{ cm}^3$) at one time, and then at a more constant, continuous level over several hours. Gas analyses were made of the coolant over this period of time, the results⁽¹⁾ being shown in Figure 4. The GOP computer program has been used to simulate this experiment and the gas compositions calculated are shown in Figure 5. A comparison of Figures 4 and 5 show them to be qualitatively very similar, especially in the $\text{H}_2/\text{H}_2\text{O}$ ratio. Quantitatively, the experimental analyses account for less than half of the injected water. It is postulated that this water may have been absorbed on some of the cooler portions of the reactor and thus removed from the gas stream. When the computer analysis is made neglecting radiation-induced reactions, a CO/CO_2 ratio of about 10 is calculated and the CO level is very close to the H_2 level. If radiation is included (as shown in Figure 4), then CO/CO_2 ratios closer to the experimental results are obtained. It is concluded that radiation-induced reactions contribute significantly to the steady-state gas compositions in this type of reactor.

The Dragon reactor injection tests have demonstrated, at least qualitatively, the usefulness of the GOP computer code. A number of other injection tests have been run at Dragon, including H_2 and CO_2 injection tests and additional H_2O tests. These tests are currently being analyzed and will be reported later.

STEADY-STATE ANALYSES IN FORT ST. VRAIN

Thermal Reactions Only

Using the steady-state calculation method outlined previously, the H_2 and H_2O concentrations and thus the $\text{H}_2/\text{H}_2\text{O}$ ratio when only thermal

FIGURE 4
Dragon Reactor Water Injection Test - August 1966

FIGURE 5
Dragon Reactor Water Injection Test - August 1966

reactions are considered is rapidly determined. The CO and CO₂ levels are also determined but somewhat less accurately.

The steady-state levels depend primarily on the water leak rate, gas purification factor, graphite temperature, gas flow rate, and graphite reactivity. All but the latter item are design parameters and are fixed for normal operation. For the graphite reactivity we have used rate constants determined for TSX graphite, which have been pre-oxidized to ~ 5% burnoff. To assess the effect of reactivity we have varied the k_1 values of equations (8) and (12) by the factors 0.1 to 10.0. The 0.1k level would be typical of a nuclear graphite which has had no oxidation. The use of ten times the rate constant is an attempt to approximate the effect of fission product impurities in the graphite which catalyze the oxidation rate. These factors are applied only to the k_1 constants of the water - graphite and carbon dioxide - graphite reactions [see Equations (8) and (12)]. Table III gives the steady-state concentrations for normal operation at the three different k levels for the thermal reactions only.

TABLE III
STEADY-STATE GAS COMPOSITIONS IN HTGR THERMAL REACTIONS ONLY

	H ₂ O	CO	CO ₂	H ₂	H ₂ /H ₂ O
0.1k	1.69×10^{-1}	1.82×10^{-1}	1.9×10^{-3}	1.91×10^{-1}	1.1
1.0k	7.72×10^{-2}	2.82×10^{-1}	6.10×10^{-4}	2.83×10^{-1}	3.7
10.0k	3.18×10^{-2}	3.27×10^{-1}	1.5×10^{-4}	3.29×10^{-1}	10.3

Effect of Radiolytic Reactions

Estimating the gamma dose rate to the gas channels to be 3.5×10^8 r/hr and to be constant over the core we have included radiation reactions and determined the steady-state, again at three different k_1 levels. The results are shown in Table IV.

TABLE IV

STEADY-STATE GAS COMPOSITIONS IN HTGR THERMAL AND RADIATION REACTIONS

	H ₂ O	CO	CO ₂	H ₂	H ₂ /H ₂ O
0.1k	8.0×10^{-2}	5.0×10^{-2}	1.5×10^{-1}	3.2×10^{-1}	4.0
1.0k	2.80×10^{-2}	1.05×10^{-1}	1.15×10^{-1}	3.25×10^{-1}	11.6
10.0k	9.0×10^{-3}	2.3×10^{-1}	6.0×10^{-2}	3.6×10^{-1}	40.0

The radiation produces a decrease in H₂O and CO concentrations and an increase in H₂ and CO₂. The overall H₂/H₂O ratio and the CO₂/CO ratio are thus increased.

Variation in Water Inleakage Rate

Calculations have been completed to determine the effect of various water leak rates on the steady-state gas composition of the HTGR. The steady-state water level is related to the leak rate as shown in Figure 6 which plots log steady-state H₂O versus log leak rate.

The time required to attain steady-state conditions varies only slightly with the leak rate. Approximately 50 percent of the steady-state water level is attained in 3-1/2 hours (reactor operating time), 80 percent in 12 hours, and 90 percent in 24 hours.

FIGURE 6
Steady State Water Level vs Leak Rate

ACCIDENT ANALYSIS IN FORT ST. VRAIN

A postulated accident for the HTGR is the rupture of a steam generator tube. It is calculated that water could leak into the reactor at the rate of 35 lbs/second for a total of 64 seconds if this occurred. This accident has been simulated on the computer.

Figure 7 shows the total steam level versus time in the primary coolant system as a result of the accident. BNW I maintains the operation of the purifier and gas circulator. In BNW II the purifier is assumed to be inoperative at the conclusion of the 64 seconds of water inleakage and the gas flow rate has been reduced to 0.1 of its original value. This latter condition simulates more realistically what would occur. The total pressure buildup for the BNW II analysis is also shown on the curve, this increase is less than 10%.

CONCLUSIONS

The GOP computer program is a useful tool for predicting steady-state impurity levels in the gas system of HTGR, and for studying the effect of a number of possible variables on those levels.

Application of the GOP code to the Dragon water injection test, while only qualitatively agreeing, does illustrate the utility of the code. For example, it was shown that radiation reactions must have had an important influence on the gas concentrations achieved.

The program was also applied to predicting operating characteristics of the Fort St. Vrain reactor.

FIGURE 7
 HTGR Accident Simulation Analysis -
 Assumed Leak 35 lbs/sec for 64 Seconds

It is calculated that a H_2/H_2O ratio of about 11 will be obtained for the projected normal operating conditions of the Fort St. Vrain HTGR, assuming graphite reactivity typical of nuclear graphite after about 5% oxidation. About 24 hours of reactor operation should be sufficient to obtain steady-state gas compositions.

The GOP program is also useful in accident analysis studies. In the hypothetical rupture of a heat exchanger a total pressure increase of less than 10% in the gas system has been calculated.

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