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Abstract

Factors influencing the possibility of a fire resulting from admission of air to the core of a high-temperature, graphite-moderated, inert gas-cooled reactor are considered. An excessive temperature rise in the core (graphite fire) may result if the rate of heat generation from internal sources exceeds the rate at which heat can be dissipated to the surroundings. Approximate calculations indicate that under several possible conditions of coolant system failure with admission of air to the core, a graphite fire could result. More accurate methods of calculation are outlined in an Appendix. Confirming experimental studies and possible methods of fighting an existing fire are proposed.

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GRAPHITE COMBUSTION HAZARD IN THE EGCR

by J. W. Prados

Introduction

Among the hazards associated with failure of one or more components of a high-temperature, inert gas-cooled power reactor is the possibility of a fire in the graphite moderator. Should rupture of coolant system circulation components occur, with resultant loss of coolant pressure and admission of air, it might be possible for the graphite oxidation to proceed sufficiently rapidly to become autothermal, i.e., the rate of heat generation by the exothermic graphite oxidation would exceed the rate at which heat could be dissipated to the surroundings. Under such conditions an excessive temperature rise in the reactor core could result in subsequent failure of fuel elements and release of fission products through the already ruptured coolant system.

The purpose of this report is to present estimates of relative rates of heat generation and dissipation so as to predict, if possible, those conditions under which an excessive temperature rise might occur, and to give possible magnitudes of the temperature rise. The model chosen for investigation was the Experimental Gas-Cooled Reactor (EGCR), now under construction at the Oak Ridge National Laboratory.

Heat Sources other than Combustion

In the event of a rupture in the coolant circulation system it is presumed that the safety system would initiate a reactor scram immediately. However, even after shut-down, heat will still be generated within the fuel and moderator, primarily from beta and gamma energy released during fission product decay. This necessitates some coolant circulation with the reactor shut down and would make inadvisable the complete isolation of the reactor core if failure should occur in both coolant lines.

Energy storage resulting from radiation damage in the graphite provides an additional heat source. Should the temperature of the moderator rise significantly, as it could on sudden reduction of coolant flow, all or part of this

stored energy could be released at a fairly rapid rate, hence aggravating any existing combustion hazard. Fortunately at the operating conditions of the EGCR the temperature of the major portion of the graphite is near 1100°F, sufficiently high that most of the stored energy will be annealed. There is a small region in the cooler portion of the core where the graphite temperature is low enough to permit accumulation of significant amounts of stored energy over periods of several years. It is anticipated, however, that procedures will be initiated at intervals to raise the temperature of the cooler portions of the core, and hence anneal most of the radiation damage before significant energy storage can occur.

Conditions of Failure Considered

One of the more difficult problems involved in any evaluation of graphite combustion hazards is prediction of the type of accident that might occur, particularly with regard to the location of ruptures in the coolant system and the extent of blower and power failures. Several possible accident conditions have been selected for consideration here. These were chosen in an attempt to illustrate a fairly wide range of conditions that might exist in the core.

a. Rupture of Coolant Line outside of Core Isolation Valves -- Both Blowers Still Operable. This is probably the least serious and at the same time most likely accident condition. By closing the block valves in the ruptured line it would be possible to isolate the core from continued introduction of fresh air, at the same time maintaining circulation with the blower in the undamaged line. Some air introduction would probably be unavoidable during the time interval between system depressurization and valve closure. However, any significant amount of oxidation would rapidly consume the available oxygen supply, thus discouraging a serious temperature rise. A previous analysis of this mode of failure (8) has predicted no serious consequences, and hence it will not be considered further in this report.

b. Rupture of Coolant Line between Isolation Valve and Core -- Both Blowers Still Operable. In the event of coolant line failure between the isolation valves and the core it would be impossible to prevent continued introduction of fresh air. Here a decision must be made whether to shut off the blower in the ruptured

line and hence reduce the amount of oxygen supplied to the core, or to keep both blowers operating so as to maintain a lower temperature in the air leaving the core.

c. Rupture at any Point Accompanied by Failure of Both Blowers. Should main and auxiliary power sources to both blowers fail simultaneously with coolant system rupture an extremely serious condition could result. Even though rupture occurred outside the block valve, it would be inadvisable to isolate the core for extended periods of time, since with no heat removal, release of decay heat can raise the core temperature at an average rate near 60°F per hour. (Local temperatures could, of course, rise considerably faster.) Maintenance of natural circulation with air at atmospheric pressure provides insufficient heat removal to prevent a serious temperature rise, even without taking into account heat release due to oxidation. Should blower failure be only temporary, it might be feasible to isolate the core until blower power could be restored. However, the longer blower power is delayed, the more likely that combustion will occur when air circulation begins.

Mechanism of Temperature Build-up

In the present analysis, specific attention was focused on the coolant channels near the center of the reactor core, since these would be subjected to the highest temperatures under normal operating conditions, and hence most susceptible to combustion. The coolant channels are formed by 5-1/4-in. diameter, 19-1/2-ft long circular holes into which are inserted the fuel element assemblies. Each assembly consists of a seven-element cluster contained inside an annular graphite sleeve of 5-in. OD and 3-in. ID. A more detailed description may be found in published design information (8). This geometry results in two parallel coolant passages per channel; an annular space outside the sleeve and a central section about the fuel element. The channels are provided with inlet orifices designed to permit approximately 6-1/2 per cent of the total flow per channel to pass through the outer annulus. This design provides essentially equal outlet temperatures for annular and central sections, since approximately 6-1/2 per cent of the total heat generation occurs in the moderator and shield and the remainder in the fuel under normal reactor operating conditions. It has been assumed in the present study that

the entire graphite sleeve has been coated with some impervious material so that only the outer wall surface of the annular space is available for oxidation.

In analyzing the possibility of autothermal oxidation on the outer surface of such a channel, one must balance the rates of heat generation from all sources against the total rate of heat loss under various conditions of operation. The heat sources are:

1. The exothermic oxidation reactions between carbon and oxygen and carbon monoxide and oxygen.
2. Delayed heat generation in the fuel elements and moderator due to fission product decay.
3. Release of stored energy due to prior radiation damage.

Heat losses from the channel surface are:

1. Conduction and convection to the flowing gas stream and thence out of the system. (Since the core is surrounded by a lagged pressure vessel, direct losses to the surroundings should be small.)
2. Endothermic reduction of carbon dioxide on the channel surface.
3. Heat conduction into the bulk of the graphite, provided that the channel surface is at a higher temperature than the remainder of the moderator.

In order to compute the rate of heat release by chemical reaction it is necessary to consider the mechanism of carbon combustion. This has been investigated extensively for a number of years (1)(2)(4)(10) but is still the subject of debate (2). A plausible mechanism consistent with observed results has been suggested as follows (2)(4).

- a. Oxygen diffuses to the carbon surface where it reacts according to

This reaction appears to be first-order in oxygen pressure and to be controlled by chemical factors below about 1340°F (2). At higher temperatures the rate of mass transfer of oxygen to the solid surface plays an important role in determining the overall reaction rate, but the rate is still lower than if mass transfer alone were controlling, at least at low gas velocities. Tomlinson (9) and Kosiba and Dienes (6) have found that previous irradiation of graphite considerably accelerates the oxidation rate at temperatures below 980°F. However, at higher temperatures, the effect of irradiation apparently becomes negligible.

b. Carbon monoxide is transferred to the bulk of the gas phase where it is oxidized

The rate of this reaction as a function of temperature and gas composition is given by Hougen and Watson (4). The reverse reaction is not important below 3000°F.

c. As carbon dioxide accumulates in the gas phase it is transferred to the wall where it is reduced at temperatures above 1300°F.

Information on the rate of this reaction is not complete, but it appears probable that mass transfer of carbon dioxide to the wall is the limiting factor at high temperatures.

As a consequence of this mechanism, one would expect a continuous decrease in oxygen concentration in the direction of flow along a coolant channel. Unless oxygen were supplied in excess, the carbon dioxide concentration would build up to a maximum and then decrease, and the carbon monoxide concentration would increase continuously until all the oxygen and carbon dioxide had been consumed. One would then expect the rate of heat release to be highest near the entrance sections of the channels where the exothermic reactions a) and b) predominate and, if the oxygen supply is limited, for there to be a cooling tendency toward the channel outlet as the endothermic reaction c) becomes more important.

Based on the above mechanism, a quantitative application of the laws of conservation of mass and energy to a differential length of channel yields a set of simultaneous partial differential equations expressing the variation in gas compositions, wall temperatures and bulk gas temperatures with time and distance along a channel. These equations may be solved numerically together with the differential equation of heat conduction in fuel, sleeve and moderator and an assumed set of initial conditions (i.e., inlet gas temperature and composition, reactor temperature and heat flux distribution, etc.). These equations are presented in the Appendix, together with suggestions for evaluating parameters which appear in them. However, their complexity and non-linear nature would make solution extremely difficult, and the time required to develop a satisfactory computer program would probably be significant. Due to these facts

and the uncertainties involved in evaluation of the reaction rate expressions appearing in the equations, it is probably not advisable for accurate numerical calculations to be attempted at this time. However, several useful approximate calculations can be made with relative ease, which will indicate under what conditions graphite combustion is likely to occur, together with the magnitude of temperature rise expected.

Approximate Calculations

In order to estimate the maximum wall temperature which the graphite can attain under oxidizing condition, a heat balance can be made about the surface of interest at a given point in the coolant channel. This balance will equate the rate at which heat is released at the wall by chemical reaction and radioactive decay to the rate at which heat is carried away into the flowing gas stream by convection. Mathematically, the balance may be represented by the following relation, based on a unit area of wall.

$$q_m + r_w (-\Delta H) = U (T_w - T) \quad (1)$$

where

- q_m = rate of decay heat release per unit area of wall, energy/area-time
- r_w = rate of oxidation reaction at wall, moles of O_2 reacted/area-time
- ΔH = heat of oxidation reaction, energy/mole of O_2 reacted
- U = overall heat transfer coefficient from oxidizing surface to bulk gas stream (includes thermal resistance of annular gap and graphite sleeve), energy/area-time
- T_w = temperature of oxidizing wall
- T = temperature of bulk gas stream

It will be noted that both sides of Equation (1) are functions of T_w . If one assumes a given set of system conditions, i.e., decay heat release rate, reaction mechanism and rate, flow conditions and gas temperature, it will be possible to solve the relation for T_w . For calculating the quantities required for such a solution, the following assumptions were made:

- a. The only reaction at the channel wall is $2 C + O_2 \rightleftharpoons 2 CO$
- b. The rate of combustion of graphite is as reported (1) and (2) but ~~in no~~ no case does it exceed the rate of mass transfer of oxygen from the bulk gas stream to the wall at the assumed flow conditions.

c. At the wall temperatures encountered, reaction rates are not appreciably accelerated by existing or previous radiation.

d. Any graphite radiation damage has been annealed, and stored energy may be neglected.

e. Heat and mass transfer coefficients are independent of temperature.

f. Steady state conditions exist in the graphite with delayed heat generation rate equal to that existing about 30 seconds after shut-down. (One third of the delayed heat generation is assumed to occur in the moderator, and to be immediately available at the wall.)

It is believed that these assumptions are conservative and will not, in general, lead to predicted wall temperatures lower than those encountered. Mass and heat transfer rates were estimated from standard correlations (5) and delayed heat generation rates from an empirical equation presented by Glasstone (3). System dimensions and operating conditions were taken from available preliminary design information on the EGCR (8). For the purpose of these calculations it was assumed that the fuel assembly graphite sleeve was coated with some impervious material and did not oxidize. Were this not the case, the calculated heat gain rates would be increased by 100-200 per cent.

The significance of the heat balance calculations can be more easily visualized if they are presented in graphical form. The heat release rate per unit area of wall is computed and plotted as a function of wall temperature, and the rate of heat loss from the wall to the flowing gas stream is plotted on the same set of axes for a given set of system conditions (gas composition, temperature and flow rate). The upper point of intersections of the two curves represents the maximum wall temperature obtainable by the graphite under steady state conditions. An increase in temperature above the intersection point would cause the heat loss rate to exceed the rate of heat release and lead to a drop in temperature back to the stable operating point. The opposite condition would obtain if the temperature tended to drop below the intersection point. The point of intersection of the heat loss curves with the horizontal axis represents the bulk air temperature at the point of interest, since if $T_w = T$, the heat loss rate is zero. The initial wall temperature will be in the neighborhood of 1100°F.

Plots of heat release rate (heat gain) and heat loss rate vs. wall temperature are presented in Figures 1 and 2. The cases illustrated correspond to

Fig.1. Relative Heat Gain and Loss at Channel Entrance.

Fig. 2. Relative Heat Gain and Loss at Channel Outlet.

hypothetical conditions of system failure b) and c) as discussed earlier in this report. Figure 1 was prepared for conditions expected near the entrance to the coolant channel; atmosphere oxygen concentration and a relatively low gas temperature. Figure 2 illustrates conditions that might be expected near the outlet end of the channels; lower oxygen concentration but higher gas temperatures. Exit gas composition and temperatures were estimated from material and energy balances on the flowing gas stream, based on assumed "average" conditions in the coolant channels. Flow rate conditions considered are as follows:

a. Both blowers operate at normal operating speed, but with mass flow rate reduced from 427,000 lbs/hr to 200,000 lbs/hr due to change of coolant from helium at 300 psia to air at 1 atmosphere.

b. Only one blower operates, and flow is poorly distributed so that circulation rate drops to 40,000 lbs/hr.

c. Flow is maintained at 1,200 lbs/hr by natural circulation.

Note that no heat loss curve is shown for natural circulation (c) in Fig. 2, since at this low flow rate, an outlet gas temperature in excess of 5000°F is calculated from the energy balance.

Results and Discussion

It can be seen from Figs. 1 and 2 that in all cases considered large temperature rises (combustion of the graphite) may be expected, even with a coating on the fuel assembly sleeve. In some instances, wall temperatures in excess of 5000°F are predicted. It should be emphasized, however, that such temperatures are highly improbable, since the carbon-oxygen reaction equilibria above 5000°F are unfavorable for the exothermic oxidation reactions. It is doubtful if temperatures as high as 5000°F could be attained, since the endothermic carbon dioxide reduction (reaction step c) has been completely neglected and would be expected to become significant at distances removed from the inlet end of the channels. For accurate calculation of maximum gas and wall temperatures, a much more detailed analysis than that presented here would be required.

An interesting comparison can be made between these results and the experimental measurements of Robinson and Taylor (7) on graphite combustion. Their system was not completely analogous to that considered here, since they attempted to maintain both graphite and air at approximately the same temperature. How-

ever, they did observe the beginning of combustion when their air and graphite temperatures exceeded about 980°F. It can be seen from Figs. 1 and 2 of this report that at this temperature the graphite oxidation rate undergoes a sudden rapid increase. If the air temperature is above this value, it is impossible for steady state to be reached (i.e., heat loss and gain rates to balance) before the graphite temperature has risen disastrously. Hence one sees the necessity of maintaining the cooling air flow sufficiently high to prevent the outlet air temperature from reaching 980°F and the inadvisability of shutting off either blower in the event of a coolant line rupture that could not be isolated from the core (failure condition b, as discussed earlier).

Future Studies Needed

It should be re-emphasized that the simplified calculations presented herein fall far short of giving a complete and accurate picture of the graphite combustion hazard in the EGCR. One means of extending knowledge in this area would be the carrying out of detailed calculations based on equations similar to those of the Appendix. A possibly more desirable approach would be to construct an experimental mock-up of a graphite coolant channel, complete with dummy fuel assemblies. Decay heat release as a function of time could be simulated by electrical resistance heating in the graphite and fuel elements. A wide range of air flow and temperature conditions could be investigated with such a device as well as the effects of graphite irradiation on the temperature rises observed.

As a long-range program of more fundamental value, particularly with regard to predicting graphite combustion hazards in future reactors, it would be highly desirable for a careful investigation to be made into the mechanisms of graphite oxidation on the inner walls of tubes. The objectives of such a study would be the identification and measurement of the rates of the individual chemical and physical steps involved in the oxidation process. With accurate rate information available, detailed calculations for graphite combustion processes would be more justifiable than they are at present.

Methods of Combating Combustion

In the event that a graphite fire should start, the question arises as to how it should be fought, so as to minimize the hazards of secondary fires and release of radioactive material to the surroundings. An obvious possibility would be to maintain on hand a large supply of some coolant that would not support combustion such as nitrogen, carbon dioxide or water. Of these, liquid water would be already available at the reactor site, and would have the most rapid quenching effect on the fire. However, the water would have to be supplied in large quantities, as small amounts might not cool the graphite rapidly enough to prevent significant hydrogen formation, which could lead to secondary fires or explosions. Also the use of a rapid water quench would probably cause severe damage to the fuel elements due to thermal shock.

Carbon dioxide and nitrogen have the disadvantage of requiring large storage facilities at the reactor site, and would hence be quite expensive to maintain in adequate supply. However, if a continuous purge of either gas could be supplied to the core immediately following system failure, it would almost certainly prevent any fire or serious temperature rise, and should not lead to any serious fuel element damage. The inert purge could be discontinued and replaced with an air flow as soon as the graphite temperature was reduced below 600-800°F. It would be highly advisable for a study to be made to determine the amount of carbon dioxide or nitrogen required for this cooling duty and the economic feasibility of such a system.

Conclusions and Recommendations

It is difficult to draw quantitative conclusions from a study of this nature since, of necessity, so much qualitative and approximate reasoning is involved. However, it is felt that the following statements can be made with a reasonable degree of confidence.

In the event of coolant system rupture during EGCR operation, it is felt that graphite combustion* will not occur if the reactor can be scrammed immediately, and one of the following two conditions is met:

* Combustion is defined here, rather arbitrarily, as a condition resulting in wall temperatures in excess of 2500°F.

a. The ruptured line can be isolated from the core and coolant circulation maintained by the blower in the undamaged line, or

b. both blowers can circulate cool air through the core at a rate of at least 200,000 lbs/hr.

For ruptures in which one of these conditions is not met, graphite combustion is highly probable. If the graphite sleeves of the fuel assemblies are left uncoated, combustion might occur even though condition b) be fulfilled.

The complete isolation of the core in the event of a simultaneous system rupture and blower failure would probably lead to a damaging temperature rise within the core due to inability to remove decay heat, even without oxidation.

It is recommended that experimental studies be carried out on a coolant channel-fuel assembly mock-up in order to define more accurately the conditions under which graphite combustion would occur. It would also be highly desirable to initiate a long-range investigation of the mechanism of graphite combustion on tube walls in order to provide more accurate rate information for calculations of graphite combustion hazards in future reactors.

It is further recommended that studies be made of the economic feasibility of maintaining a supply of some inert gas at the reactor site, to be used as an emergency coolant in the event of system rupture until the graphite temperature could be reduced below ignition levels.

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Appendix

The following system of equations is believed to provide a reasonably accurate representation of temperature-composition-time-distance relations for the model under investigation. Major assumptions involved in their derivation are:

1. Gas flowing in main coolant channel and annulus is perfectly mixed in radial direction (except for a thin film at the walls) and unmixed in the longitudinal direction.
2. Longitudinal heat conduction along coolant channels is negligible.
3. Thermodynamic properties of all components are additive.
4. Radial symmetry exists in the system.
5. No radiant heat exchange occurs between outer channel wall, sleeve and fuel elements.
6. Fuel assembly graphite sleeve is coated to prevent oxidation.

Notation employed in these equations is presented at the end of the Appendix.

Material Balances (Annulus)

$$\frac{S_A}{a_{wo}} \frac{\partial \rho_P}{\partial t} + \frac{1}{a_{wo}} \frac{\partial N_P}{\partial z} = -r_a + \frac{S_A}{a_{wo}} r_b \quad (2)$$

$$\frac{S_A}{a_{wo}} \frac{\partial \rho_Q}{\partial t} + \frac{1}{a_{wo}} \frac{\partial N_Q}{\partial z} = 2r_a - 2\frac{S_A}{a_{wo}} r_b + 2r_c \quad (3)$$

$$\frac{S_A}{a_{wo}} \frac{\partial \rho_R}{\partial t} + \frac{1}{a_{wo}} \frac{\partial N_R}{\partial z} = -2\frac{S_A}{a_{wo}} r_b - r_c \quad (4)$$

$$\frac{S_A}{a_{wo}} \frac{\partial \rho_I}{\partial t} + \frac{1}{a_{wo}} \frac{\partial N_I}{\partial z} = 0 \quad (5)$$

heat capacities can be computed from available thermodynamic data. Heat and mass-transfer coefficients may be obtained from standard correlations (5), and rates of decay heat release may be estimated as functions of time from the equation given by Glasstone (3). Initial conditions are provided by the assumed conditions of failure, and boundary conditions are the equality of temperatures and heat fluxes at the fluid solid interfaces.

Gas-Phase Energy Balance (Annulus)

$$\begin{aligned}
 S_A \bar{\rho} \bar{C}_P \frac{\partial T_A}{\partial t} + N_{TA} \bar{C}_P \frac{\partial T_A}{\partial z} = h_{wo} a_{wo} (T_{wo} - T) \\
 - h_{wi} a_{wi} (T_A - T_{wi}) + S_A r_b (-\Delta H_b) + r_a a_{wo} \int_{T_A}^{T_{wo}} (2C_{PQ} - C_{PP}) dT \\
 + r_c a_{wo} \int_{T_A}^{T_{wo}} (2C_{PQ} - C_{PR}) dT
 \end{aligned} \quad (6)$$

Gas-Phase Energy Balance (Central Channel)

$$S_e \bar{\rho} \bar{C}_P \frac{\partial T_C}{\partial t} + N_{TC} \bar{C}_P \frac{\partial T_C}{\partial z} = h_{wC} a_{wC} (T_{wC} - T_C) + h_f a_f (T_{wf} - T_C) \quad (7)$$

Heat Conduction in Moderator, Sleeve and Fuel Elements

$$\frac{\partial T_M}{\partial t} = \alpha_M \nabla^2 T_M + \frac{Q_M}{\rho_m C_{p_m}} \quad (8)$$

$$\frac{\partial T_S}{\partial t} = \alpha_s \nabla^2 T_s + \frac{Q_s}{\rho_s C_{p_s}} \quad (9)$$

$$\frac{\partial T_F}{\partial t} = \alpha_f \nabla^2 T_f + \frac{Q_f}{\rho_f C_{p_f}} \quad (10)$$

Heat Balance at Outer Wall of Annulus

$$-k_m \left. \frac{\partial T_M}{\partial n} \right|_{n=n_{wo}} + h_{wo} (T_{wo} - T_A) = r_a (\Delta H_{aw}) + r_c (-\Delta H_{cw}) \quad (11)$$

It may be desirable to consider the heterogeneous nature of the fuel element-can assembly and apply Equation (10) to two separate regions.

The rate expressions appearing in Equations (2) - (11) may be evaluated, once conditions of operation, i.e., flow rate, initial composition, etc. corresponding to a given type of failure have been assumed (1) (2) (4). Heats of reaction and

Notation

C_p	Molal heat capacity, energy/mole-degree
ΔH	Heat of reaction, energy/mole
N	Molal flow rate of gas, moles/time
Q	Heat generation rate per unit volume, energy/volume-time
S	Cross sectional area of channel available for flow, area
T	Temperature, degrees
U	Overall heat transfer coefficient, outer annulus wall to bulk gas stream, energy/area-time-degree
a	Wall surface area per unit channel length
h	Surface heat transfer coefficient, energy/area-time-degree
k	Thermal conductivity, energy/length-time-degree
n	Radial coordinate in channel, length
q	Heat generation rate per unit channel wall area, energy/area-time
r	Reaction rate, moles/area-time or moles/volume-time
t	Time
z	Longitudinal coordinate along channel, measured from inlet end, length
α	Thermal diffusivity, length ² /time
ρ	Molal density, moles/volume
∇^2	Laplacian Operator, $(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2})$ in cartesian coordinates

Subscripts

A	Annulus condition
C	Central channel condition
I	Inert gas, e.g., N ₂ , He

M	Condition in moderator
P	Oxygen
Q	Carbon monoxide
R	Carbon dioxide
s	Condition in sleeve
T	Total gas stream
W	Condition at fluid-solid interface
a	Reaction step $2\text{ C} + \text{O}_2 \rightleftharpoons 2\text{ CO}$
b	Reaction step $2\text{ CO} + \text{O}_2 \rightleftharpoons 2\text{ CO}_2$
c	Reaction step $\text{CO}_2 + \text{C} \rightleftharpoons 2\text{ CO}$
f	Condition in fuel element
i	Inner wall of annulus
o	Outer wall of annulus