SIMULATION OF THE COMBUSTION CHAMBER OF A H_2-O_2 THERMAL RECOMBINER

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ABSTRACT

Thermal recombiners are installed in many nuclear power plants to prevent late hydrogen deflagrations during a design basis accident (DBA); experimental tests guarantee DBA operation. A code has been made to simulate the steady-state operation of the FIAT recombiner. The recombining rate, calculated with the code for a gas mixture typical of severe accidents, is higher than the highest value obtained in the DBA experimental tests. This result is encouraging for the use of thermal recombiners even in severe accidents, but the recombiner capability and stability should be verified through experimental tests.

1.0 INTRODUCTION

In a loss of coolant accident (LOCA) in a water-cooled nuclear power plant, some amounts of hydrogen, even considerable, can be produced and released together with steam into the safety containment. Hydrogen is produced very fast by a metal-water reaction when, at the beginning of the accident, zirconium and steel at a high temperature come in contact with steam. Other slower reactions producing hydrogen are water radiolysis and aluminium corrosion.

The gas mixture composition in the containment depends on the containment free volume and gas mixing rate, as well as on the amount of hydrogen released. If the molar fractions of hydrogen and oxygen exceed the respective flammability limits, 4-5% for H_2 and 5-6% for O_2, a very small ignition source can cause the rapid propagation of a combustion wave [1]. A subsequent rise in pressure or even shock waves can damage the containment structure. A high steam concentration increases the flammability limits. Generally, the gas mixture is never flammable when steam concentration is more than 50-60% in volume.

To prevent hydrogen explosions in the safety containment, the reactants could be recombined in a controlled manner to

counterbalance the reactant production rate before flammability is reached. Among the various types of recombiners that are on the market or that have been proposed, thermal recombiners installed outside the containment can be well controlled and easily replaced with backup units.

In pre- or post-inerted containments, a thermal recombiner can eliminate the oxygen produced by radiolysis to keep oxygen below its flammability limit in any accident.

Thermal recombiners for large non-inerted containments were designed so that only one unit is needed to keep hydrogen below its flammability limit during a design basis accident (DBA). In this case, the hydrogen generated by metal-water reaction is limited, because of the drop in temperature due to the emergency core cooling systems, and flammability would be reached after a few days, when hydrogen is slowly produced by water radiolysis and aluminium corrosion.

In a severe accident, the gas mixture becomes flammable early, when the hydrogen production rate is still great. Generally, in this type of accident, an impractical number of thermal recombiners would be needed to prevent combustion*, but a non-excessive number could mitigate the combustion overpressure by reducing the reactant concentrations. Therefore, it is important to assess the performance of the recombiner in a severe accident, when hydrogen and steam concentrations are higher than in a DBA.

2.0 THE FIAT-CIEI THERMAL RECOMBINER

The thermal recombiner constructed by FIAT-CIEI [4], on a Rockwell design [5], is installed outside the nuclear reactor safety containment and may be easily transported, so it can also be used as a backup hydrogen or oxygen control system.

The gas is sucked from the containment, pre-heated by electrical heaters, sent to a chamber (an insulated cylinder with vertical axis) where the combustion takes place and, after cooling, brought again into the containment (Fig. 1).

Actual design pressure is 415 kPa gauge [4]. Actual maximum operating temperature of the inlet gas is 355 K due to the working limit of the blower motor [4], but it could be increased. In the reaction chamber, the outlet gas temperature reaches the steady state in 1-2 hours and is kept close to a pre-fixed value of 988 K by a special control system that regulates the electrical power of the radiant elements. The chamber design

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* In some severe accident, a very high concentration of steam could inert the containmemt atmosphere long enough to allow a non-excessive number of recombiners to reduce the concentration below its flammability limit before steam condensation [2, 3].

temperature is 1050 K. Between the inlet and the outlet gas in the reaction chamber, the rise in temperature is proportional to the drop in the molar fraction of hydrogen: 60-70 K for each %. Flammable mixtures with H_2 concentrations up to 10% in volume cannot be ignited by the combustion in the reaction chamber, because no flashback is possible below this concentration while the blower is working [6].

Acceptance tests are carried out under the same inlet gas conditions as those in the containment atmosphere when the recombiner operates due to design basis LOCAs. In these tests, the hydrogen concentration in the outlet gas flow is below 0.1% in volume (Tab. 1).

Table 1
Range of Inlet Gas Conditions that Guarantee Outlet Hydrogen Molar Fraction Below 0.1%.

	Min.	Max.
Pressure (kPa)	101	150
Temperature (K)	290	355
Flow rate ($STPm^3/h$)	119	153
Humidity (%)	0	100
Mixture composition (% in vol.)		
hydrogen	0.5	5
oxygen	15	20
nitrogen	75	80
water drops		$5\ 10^{-5}$

In severe accidents, some devices could modify the physical and chemical properties of the gas sucked from the containment, so the gas inlet in the reaction chamber is the same as in acceptance tests:
- a recirculation loop might dilute the gas mixture, sucked from the containment, with the outlet burnt gas [6];
- a water drop separator or a steam condenser might be installed on the suction line.

However, before using this recombiner in severe accidents, working characteristics of the recombiner should also be assessed under conditions differing from those of the acceptance tests to ensure safety and/or to design suitable recirculation and condensation systems.

A computer code was created i) to determine whether an expensive experimental program should be carried out in order to investigate the whole range of the recombiner performances and ii) to identify the most suitable test conditions. In order to simulate the steady operation of the combustion chamber, the mutual influence among chemical, thermal and fluid dynamic quantities was studied and outlined in mathematical terms.

3.0 MODEL OF REACTION KINETICS

The reaction between hydrogen and oxygen is a chain-branching process that moves from the starting reactants to the final products through a series of consecutive, competitive and opposite reactions [7-12]. Unstable intermediate chemical species, sometimes very reactive (like atoms or radicals), partake in the different reactions and are either destroyed or created so that the chain is sustained independent of the reaction which started the process. The overall reaction rate depends on the concentration of these intermediate species during the evolution of the chemical transformation. In some reactions of the combustion process, more radicals are formed than react. Two regimes can be identified: an "explosive" one and a "non-explosive" one. In the non-explosive regime, the concentration of an intermediate species reaches a steady state, based on the concentrations of the "stable" reactants.

Experimentally found T, P conditions that separate non-explosive from explosive regions form a reversed S-curve [8] for each H_2-O_2 composition of an unburnt mixture. For each pressure, a temperature exists above which the mixture is explosive, and within a certain range of temperatures three different values for the explosive limit pressure exist. The working pressures of the recombiner are certainly higher than the lower limit of explosion. Hence, the reaction model must be able to explain the second and the third explosion limits.

Among the large number of reactions that are considered for the hydrogen-oxygen system, those chosen for the combustion mechanism in the recombiner are:

j)	$H_2 + O_2 \xrightarrow{k_j} HO_2 + H$		initiation
1)	$H_2 + OH \xrightarrow{k_1} H_2O + H$		propagation
2)	$H + O_2 \xrightarrow{k_2} OH + O$		branching

3)	$O + H_2$	$\xrightarrow{k_3}$	$OH + H$	branching
4)	$O + H_2O$	$\xrightarrow{k_4}$	$OH + OH$	branching
6)	$H + O_2 + M$	$\xrightarrow{k_6}$	$HO_2 + M$	prop.-term.
11)	$HO_2 + H_2$	$\xrightarrow{k_{11}}$	$H_2O_2 + H$	propagation
t)	$H + OH + M$	$\xrightarrow{k_t}$	$H_2O + M$	termination
d1)	H	$\xrightarrow[wall]{k_{d_1}}$	$1/2\ H_2$	destruction
d2)	O	$\xrightarrow[wall]{k_{d_2}}$	$1/2\ O_2$	destruction
d3)	OH	$\xrightarrow[wall]{k_{d_3}}$	$1/2\ H_2O + 1/4\ O_2$	destruction
d4)	HO_2	$\xrightarrow[wall]{k_{d_4}}$	$1/2\ H_2O + 3/4\ O_2$	destruction
d5)	H_2O_2	$\xrightarrow[wall]{k_{d_5}}$	$H_2O + 1/2\ O_2$	destruction

Each single rate of wall destruction was assumed proportional to the concentration of the intermediate species, with an unknown proportionality constant [8]. This constant was the same for all ($k_{di} = k_d$) since it essentially depends on the convective transport mechanism which is the same for all the chemical species. k_d is a parameter still to be determined through best fit analysis of experimental and computational data.

The semi-empirical relationships suggested by Baulch et al. [13] and recently confirmed [14], were used to express the other rate constants, k_i, as a function of gas temperature within the reaction volume.

In the combustion chamber of the recombiner, the regime of the reaction should be non-explosive, as the operating experience suggests; so, the local hydrogen burning rate, r_f, is an exclusive function of k_d, temperature and the H_2, O_2, N_2, H_2O

concentrations. The burning rate increases as the temperature and deficiency reactant concentration increase and k_d decreases.

Introducing other reactions into the combustion mechanism can lead to physically unacceptable results (for example, negative concentrations of intermediate species) or more than one acceptable value for the burning rate [15].

In conclusion, we chose the most complete reaction mechanism among those with congruent physical-chemical hypotheses and a sure, univocal solution.

4.0 CHEMICAL REACTOR MODEL

Once the relationship that links the local burning rate to thermodynamic co-ordinates has been found, the distribution of these co-ordinates in the reaction volume must be specified. In other words, the most suitable chemical reactor model must be chosen.

4.1 Ideal Chemical Reactors

A constant volume chamber is considered, which is thermally insulated from the surroundings and has an inlet and outlet duct. The mixture of chemical reactants is let in with a constant mass flow rate and a given temperature and pressure. The exothermic reaction takes place in the chamber and the pressure is uniform in the system.

If the inlet fluid is not mixed with the content of the chamber ("plug-flow" reactor), the conversion of reactants into products along with the fluid temperature vary continuously within the reactor.

On the contrary, if the chamber geometry can assure an instantaneous mixing of the inlet flow with the content of the reactor (perfectly "mixed-flow" or "stirred" reactor), the temperature and the concentration will be uniform throughout the whole volume and, thus, the outlet gas will have the same thermodynamic conditions as the mixture inside the chamber.

Both reactor models can be mathematically solved by mass and energy balances, once the inlet conditions and the reaction kinetics are known [16, 17] and the pressure is assumed uniform:

plug-flow reactor

$$\frac{d\eta}{dV} = \frac{r_f(\eta, T)}{J_{in}\, y_{f,in}}$$

$$\frac{dT}{dV} = \frac{r_f(\eta, T)\ \Delta h_r(T)}{J(\eta)\ C_p(\eta, T)}$$

stirred reactor

$$\frac{V}{y_{f,in} J_{in}} = \frac{\eta_{out}}{r_{f,out}}$$

$$J_{in} \int_{T_{in}}^{T_{out}} C_{p,in} dT = r_{f,out} \Delta h_{r,out} V$$

For a perfectly-mixed-flow reactor, one or three solutions to the system of equations may be found, depending on the boundary conditions:

- in one case, the solution defines a stable operation and may correspond to a "reactor out" (relatively low inlet temperature and relatively high flow-rate/volume ratio) or to a high degree of reactant conversion (relatively high inlet temperature or relatively low flow-rate/volume ratio);
- in the other case, only one solution corresponds to an effective stable reaction rate*: the one that involves the highest degree of reactant conversion and the highest outlet temperature [16, 17, 18]. Only for these values would an infinitesimal increase in conversion cause a negative feedback, because the consequent rise in outlet temperature would increase the energy removed by the gas flow more than the energy released by the reaction [17].

Instability cannot exist in the plug-flow reactor, since the reaction rate does not depend on the outlet conditions.

4.2 Real Reactor Model

In a real reactor, the mix of entering fluid with the fluid present inside the reactor is neither entirely absent (like in the plug-flow reactor) nor ever complete (like in the stirred reactor).

An increase of mixing tends to homogenize all the parameters, both physical and chemical, and make them closer to the outlet conditions. When mixing increases, the temperature rise tends to increase the burning rate but the reactant concentration drop tends to decrease it.

Experience shows that the reaction rate can exceed all predictions because the temperature is generally more uniform than the chemical composition ("segregation" phenomenon) [16, 19].

To assess the recombiner burning rate, the model need not describe exactly the actual distributions of chemical and physical quantities in the reactor.

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* The other solutions correspond to a reactor out and to an unstable operation.

To simulate the reaction volume of the recombiner, a plug-flow reactor model with a recirculation loop was chosen. A fraction of the outlet gas is recirculated and perfectly mixed with the inlet gas before entering the reactor again. The intermediate situations between plug-flow and perfectly-mixed-flow are covered by varying the recirculated fraction of the flow rate, RICIN, from 0 to 1.

The k_d value (AKD in the code) determined through best fit analysis of experimental and computational data will be affected by the difference between the temperature-chemical composition coupling of the reactor model and the real case. The closer to reality the reactor scheme is, the closer k_d will be to the original physical value and the less deformed will be the chemical kinetics. This demonstrates that RICIN is needed to reduce the difference between reactor model and actual reactor.

A plug-flow reactor with a recirculation loop shows stability problems similar those of a stirred reactor, because inlet gas is affected by outlet gas. The reaction rate at any point of the partially mixed actual reactor is affected by the upstream and downstream conditions in a way that certainly differs from the "recycle" plug-flow reactor. Even if the model can provide the actual burning rate, it cannot be used to judge actual stability, but the real reactor is more likely to be stable if the model is stable.

5.0 THERMOHYDRAULIC MODEL

The burning rate depends on the reactor thermohydraulics and, in turn, fluid pressure, temperature and velocity distributions in the combustion chamber are determined by the chemical reaction rate and the combustion heat.

The thermohydraulic model describes how, in the sections that divide the combustion chamber into zones (Fig. 2), the thermohydraulic quantities depend on the degree of overall reactant conversion, H, and on the inlet or outlet gas conditions.

Until now experimental tests have only been aimed at assessing the conversion degree of the combustion chamber as outlet gas temperature, feed flow and gas mixture composition vary. Pressure, temperature and gas speed have not yet been measured in the various chamber sections, so the irreversibility parameters (for example, friction factors) cannot be assessed.

To compensate for the missing information, a series of conditions were deduced by qualitative considerations about the actual behaviour of the inner thermohydraulics. The problem was solved by mass, energy and momentum balances between the downstream and upstream sections of each chamber zone.

We assumed the following:
- ideal gas (validity of ideal gas equation of state, specific heat capacities dependent only on temperature);
- adiabatic system, because it is thermally insulated and placed in an electrically heated environment;
- reaction volume restricted to a zone of the combustion chamber (Fig. 2);
- no friction along the walls;
- the same pressure in all the sections at the same axial position in the combustion chamber scheme (Fig. 2: $P_A = P_G$, $P_1 = P_{1'}$, $P_2 = P_U = P_F$);
- $P_W = P_{U'}$, $T_W = T_U$;
- $T_2 = T_U$ *.

The fluid conditions at the combustion chamber outlet were then linked to the recombiner outlet conditions (sections E and SC, Figs. 1 and 2) through a simulation of the return pipe made under the following hypotheses:
- constant heat capacity of the gas;
- pressure drop in the pipe due to friction alone with an estimated constant friction factor (very turbulent flow);
- subdivision of the pipe into ten parts: in each one, temperature is constant and equal to the average temperature as calculated assuming a linear variation of temperature along the pipe.

The equation system can be solved with a trial and error method, starting from a guess value of $(T_E - T_{SC})$.

6.0 THE CODE TREE

To assess the performances of the combustion chamber, semi-empirical relationships that link the unknown parameters AKD and RICIN with quantities which influence the mixing in the reaction volume must be included in the mathematical model. These relationships may be found by comparing model predictions with experimental data. For this purpose, the code TREE (Thermal Recombiner Efficiency Evaluation) was made so that for each experimental test and for each value of RICIN given in input, it gives the value that AKD must assume to obtain the experimental degree of overall conversion. For each experimental test, an iso-conversion curve can be drawn in the (RICIN, AKD) plane. The wider the range of thermohydraulic situations experimentally investigated, the greater the possibility to determine how the trend of the iso-conversion curves changes as a function of the quantities which characterize the process.

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* The code for the various experimental tests (see further on) yielded a difference of a few hundredths degrees between T_2 (= T_U) and T_F. The chosen model should be equal to those based on the hypothesis $T_F = T_U$ instead of $T_2 = T_U$ or $P_U = P_F$.

6.1 Available Experimental Data

FIAT-CIEI carried out six tests to verify the steady state behaviour of the recombiner for limited variations of the feed gas composition (without steam) and of the control parameters [20]. The number of tests and the range of investigated thermohydraulic situations are very limited and uncertainties about measurements exist, as shown in Table 2 for the measurements of the outlet volumetric concentration.

Table 2
FIAT Experimental Data

	INLET				OUTLET		
TEST	J_A mol/s	H_{2A} % vol.	O_{2A} % vol.	N_{2A} % vol.	P_{AMB} Pa	H_2 % vol.	T_U K
F1	1.3641	1.07	20.78	78.15	99700	< 0.01	993
F2	1.3641	3.08	20.35	76.57	98900	< 0.01	988
F3	1.0541	4.00	20.16	75.84	98900	0.01-0.1	1003
F4	1.5997	4.00	20.16	75.84	98900	0.01-0.1	993
F5	1.3641	4.10	20.14	75.76	99200	< 0.01	993
F6	1.3641	5.02	19.95	75.03	98900	< 0.01	988

By comparing the experimental data of tests F1, F2, F5 and F6 with the same feed flow rate (110 STP m^3/h), we find that when the inlet concentration of the deficiency reactant (hydrogen) increases, so does the degree of overall conversion and the burning rate. A slight variation in the temperature at the combustion chamber outlet does not modify this trend.

When we compare experimental data of tests F3, F4 and F5 we find that under almost the same feed chemical composition, a flow rate increase (129 STP m^3/h) or decrease (85 STP m^3/h) with respect to the value of 110 STP m^3/h leads to a decrease in the degree of overall conversion. This is not counterbalanced in the latter case by a slight increase in temperature at the combustion chamber outlet. This fact (measurement errors excepted) demonstrates that an intermediate value of flow is optimal, at least in the range of experimental conditions we examined. The effect of the flow rate increase may be due to the decrease in permanence time of the gas in the reactor and to the increase in radical destruction. The effect of the flow rate decrease may be due to the dominant effect of a decrease in the average temperature in the reactor (for the decreased mixing) not counterbalanced by the limited increase of the outlet temperature.

6.2 Mathematical Formulation of the Code TREE

The code TREE has as inputs the experimental data:

$$J_A,\ y_{A,i},\ T_U,\ T_{SC},\ P_{AMB},\ H.$$

The thermohydraulic quantities to be calculated are:

$$P_A,\ P_U,\ P_1,\ P_V,\ P_M,\ P_Z,\ P_E,\ T_A,\ T_F,\ T_G,$$

$$T_1,\ T_{1'},\ T_Z,\ T_E,\ J_F,\ J_G,\ J_1,\ J_{1'},\ J_2,\ J_U,\ y_{1,i}.$$

In the reaction volume, the chemical reactor model assumes a uniform pressure ($P_1 = P_{1'}$) and gives a virtual temperature, T_{1v}, to the inlet fluid instead of considering the variation of kinetic energy [15].

Code steps:
1) Read the experimental data, the pre-fixed value for RICIN and a trial value for AKD.
2) Calculate P_E by a trial and error method.
3) Calculate thermohydraulic quantities in the combustion chamber sections.
4) Calculate T_{1v}.
5) Calculate input conditions for the plug-flow reactor.
6) Calculate a new value for H.
7) Compare the calculated value H with the experimental value. If the relative error is greater than a pre-fixed value, vary AKD and return to 6); otherwise, print the results and stop.

6.3 Results of TREE for the FIAT Tests

For each experimental test, an iso-conversion AKD vs. RICIN curve was drawn on the basis of the values computed by the code (Fig. 3). If an increase of RICIN (i. e. of turbulence and mixing) tends to increase/decrease H, AKD must increase/decrease to counterbalance the RICIN effect.

The thermohydraulic situation joined to each iso-conversion curve is in Table 3.

By examining each curve we find:
- An increase of RICIN involves a rise in temperature and a decrease in reactant concentration in the gas entering the plug-flow reactor. When the inlet gas has a relatively low temperature and a relatively high reactant concentration (relatively high burning rate, tests F2-F6), AKD rises with mixing at low and intermediate RICIN values, to counterbalance the rise in burning rate due to the prevalent effect of the rise in temperature, and decreases slightly at high RICIN values to counterbalance the prevalent effect of the drop in reactant concentration. In test F1, AKD remains nearly constant because the feed mixture, independently of the fictitious recirculation, already has a relatively low hydrogen

Table 3

Results of the Code TREL for the FIAT Tests

Variables	TESTS F1	F2	F3	F4	F5	F6
H	0.9907	0.9968	0.9755	0.9755	0.9976	0.9980
P_V (Pa)	102916	102017	100763	103122	102265	101912
P_M (Pa)	102952	102047	100779	103158	102291	101934
P_A (Pa)	102810	101931	100715	103015	102189	101845
T_A (K)	911.812	751.982	702.325	691.252	676.563	597.151
v_A (m/s)	56.053	46.626	34.057	49.735	41.840	37.057
P_1 (Pa)	102902	102006	100756	103107	102254	101902
T_1 (K)	951.131	860.612	837.541	826.806	817.796	765.464
T_{1v} (K)	951.579	860.949	837.727	827.204	818.081	765.700
v_1 (m/s)	32.396	28.124	20.941	30.609	25.883	23.551
$y_{1,H2}$ (%)	0.56261	1.69278	2.28577	2.28927	2.31918	2.92986
$y_{1,O2}$ (%)	20.5779	19.7889	19.4617	19.4631	19.4140	19.0919
$y_{1,N2}$ (%)	78.3499	77.1094	76.5033	76.5019	76.4487	75.8343
$y_{1,H2O}$ (%)	0.51012	1.40892	1.74921	1.74564	1.81809	2.14396
$P_{1'}$ (Pa)	102902	102006	100756	103107	102254	101902
$T_{1'}$ (K)	993.006	988.006	1003.00	993.007	993.005	988.005
$v_{1'}$ (m/s)	4.291	4.073	3.156	4.626	3.952	3.811
$y_{1',H2}$ (%)	0.01000	0.01001	0.09995	0.09995	0.01005	0.01030
$y_{1',O2}$ (%)	20.3579	19.1082	18.5713	18.5713	18.4727	17.8932
$y_{1',N2}$ (%)	78.5664	77.7637	77.3491	77.3491	7.34177	76.9578
$y_{1',H2O}$ (%)	1.06570	3.11801	3.97964	3.97964	4.17555	5.13868
P_2 (Pa)	102900	102.003	100755	103104	102252	101900
T_2 (K)	993.000	988.000	1003.00	993.000	993.000	988.000
v_2 (m/s)	5.772	5.479	4.245	6.222	5.316	5.126
P_U (Pa)	102900	102003	100755	103104	102252	101900
T_U (K)	993.000	988.000	1003.00	993.000	993.000	988.000
v_U (m/s)	7.966	7.915	6.259	9.190	7.894	7.845
P_F (Pa)	102900	102.003	100755	103104	102252	101900
T_F (K)	992.977	987.977	1002.99	992.969	992.977	987.977
v_F (m/s)	7.368	6.640	5.017	7.339	6.228	5.772
P_G (Pa)	102810	101931	100715	103015	102189	101845
T_G (K)	992.776	987.816	1002.89	992.774	992.837	987.857
v_G (m/s)	22.789	20.534	15.511	22.698	19.258	17.848
P_Z (Pa)	102903	102007	100757	103108	102255	101903
T_Z (K)	993.027	988.026	1003.02	993.035	993.026	988.025
P_E (Pa)	102448	101563	100489	102509	101817	101472
T_E (K)	991.926	986.953	1002.35	991.595	991.966	986.985
P_{SC} (Pa)	99700	98900	98900	98900	99200	98900
T_{SC} (K)	298.150	298.150	298.150	298.150	298.150	298.150

concentration and a relatively high temperature.
- The slopes of the curves become steeper above a certain temperature of the gas entering the plug-flow reactor (940 K [15]), because the rates of the propagation reactions tend to rise much faster than at lower temperatures.

By comparing the curves, we observe that:
- In the experimental tests F1, F2, F5, F6, although the fluid dynamic regime is the same in the reaction volumes*, the curves shift towards lower values of AKD when the difference in the concentrations of the deficiency reactant between the inlet and the outlet is greater. In this case, the "segregation" effect should be greater.
- Following a lower mixing, caused by a lower flow rate, the curve F3 shifts towards lower RICIN values than the curve F5, which has the same feed mixture composition.
- Following a greater destruction rate of radicals, caused by a greater flow rate, the curve F4 shifts towards higher AKD values than the curve F5, which has the same feed mixture composition.

7.0 DEGREE OF CONVERSION IN SEVERE ACCIDENT STEADY-STATE OPERATION

From the analysis of the iso-conversion curves F1, F2, F5, F6, a procedure was devised to predict the conversion degree for new operating conditions, unlike the experimental ones for the gas chemical compositions at the reaction volume inlet.

We assumed that:
- an "optimum" value of RICIN characterizes entirely the reaction volume fluid dynamics;
- the AKD value, which must be utilized in TREE with the "optimum" RICIN value, is an exclusive function of the difference in deficiency reactant concentration at the reaction volume inlet and outlet, even when the feed gas mixture has a high percentage of steam.

We estimated the recombining capability when the feed gas mixture has a high hydrogen concentration and enough steam to make the mixture practically non-flammable. The new operating condition has the same fluid dynamic regime in the reaction volume and the same "optimum" RICIN value as the tests F1, F2, F5, F6. The recombiner feed flow was higher than the one used in the above mentioned FIAT tests to obtain nearly the same value of the Reynolds number (about $5\ 10^3$) at the reaction volume outlet.

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* To compare qualitatively the levels of reactor turbulence and mixing in two tests, the ratio between the respective Reynolds numbers, computed at the reaction volume outlet, was taken as a reference.

The considered mixture has the following characteristics:

feed flow:	137 STP m^3/h,
feed mixture composition:	% H_2 = 10, % air = 40, % steam = 50,
reaction volume outlet temperature:	990 K,
containment pressure: (within the limits of the DBA recombiner operation)	150 kPa,
containment temperature: (saturation temperature)	365 K.

These conditions might occur in a severe accident
- in the full-pressure containment of a pressurized water reactor of Western design (PWR) or Russian design (WWER 1000), after an early venting of incondensable gases;
- in the room where the reactor vessel of a Western BWR or a Russian pressurized water reactor WWER-440/213 [2] is placed, after part of the incondensable gases has been discharged into the downstream rooms of a pressure suppression system.

For each RICIN value, the values $K_j = \frac{AKD_{F1}}{AKD_j}$ computed for the experimental tests were plotted as a function of $Y_j = \left[\frac{(y_1 - y_{1,})_j}{(y_1 - y_{1,})_{F1}}\right]_{H_2}$, as well as the K values which were computed with TREE for the new mixture by assuming different degrees of overall conversion. In this way, two different curves, the former "semi-empirical" and the latter "theoretical", can be drawn (Fig. 4). Their intersection point on the theoretical curve determines the H value which, for the new mixture and for the pre-fixed RICIN value, satisfies the experimentally found link between AKD and the difference in deficiency reactant concentration at the reaction volume inlet and outlet.

The pairs of K and Y values were plotted for RICIN values between 0.1 and 0.9 (the reaction volume is neither a plug-flow reactor nor a perfectly mixed flow reactor). Figure 4 shows these plots for RICIN = 0.5.

Table 4 shows the values of the thermohydraulic quantities in the combustion chamber which were calculated for different assumed H values.

Table 4

Results of the Code TREE for the New Mixture

Variables		ASSUMED CONVERSION DEGREE					
H		0.2	0.3	0.4	0.5	0.6	0.7
P_V	(Pa)	152032	152012	151992	151973	151953	151933
P_M	(Pa)	152058	152036	152014	151991	151969	151947
P_A	(Pa)	151956	151942	151929	151916	151903	151891
T_A	(K)	852.912	782.914	711.573	638.799	564.520	488.730
v_A	(m/s)	44.210	40.585	36.890	33.120	29.272	25.344
P_1	(Pa)	152023	152003	151984	151964	151945	151926
T_1	(K)	917.901	879.069	837.771	793.297	744.751	691.109
T_{1v}	(K)	918.088	879.235	837.916	793.420	744.854	691.193
v_1	(m/s)	25.820	24.166	22.455	20.659	18.766	16.776
$y_{1,H2}$	(%)	9.10278	8.68336	8.28995	7.93142	7.61870	7.36377
$y_{1,O2}$	(%)	7.96745	7.76524	7.57557	7.40273	7.25196	7.12906
$y_{1,N2}$	(%)	31.7492	31.8190	31.8844	31.9440	31.9960	32.0384
$y_{1,H2O}$	(%)	51.1805	51.7324	52.2501	52.7218	53.1333	53.4687
$P_{1'}$	(Pa)	152023	152003	151984	151964	151945	151926
$T_{1'}$	(K)	990.003	990.003	990.003	990.002	990.002	990.002
$v_{1'}$	(m/s)	3.524	3.434	3.338	3.233	3.117	2.991
$y_{1',H2}$	(%)	8.08081	7.10660	6.12245	5.12821	4.12371	3.10881
$y_{1',O2}$	(%)	7.47475	7.00508	6.53061	6.05128	5.56701	5.07772
$y_{1',N2}$	(%)	31.9192	32.0812	32.2449	32.4103	32.5773	32.7461
$y_{1',H2O}$	(%)	52.5252	53.8071	55.1020	56.4103	57.7320	59.0674
P_2	(Pa)	152021	152001	151.982	151963	151943	151924
T_2	(K)	990.000	990.000	990.000	990.000	990.000	990.000
v_2	(m/s)	4.740	4.619	4.490	4.348	4.193	4.024
P_U	(Pa)	152021	152001	151982	151963	151943	151924
T_U	(K)	990.000	990.000	990.000	990.000	990.000	990.000
v_U	(m/s)	6.668	6.635	6.602	6.569	6.536	6.503
P_F	(Pa)	152021	152001	151982	151963	151943	151924
T_F	(K)	989.989	989.989	989.989	989.989	989.989	989.988
v_F	(m/s)	5.925	5.636	5.326	4.982	4.601	4.184
P_G	(Pa)	151956	151942	151929	151916	151903	151891
T_G	(K)	989.905	989.913	989.921	989.929	989.937	989.945
v_G	(m/s)	18.319	17.426	16.466	15.402	14.223	12.934
P_Z	(Pa)	152023	152004	151985	151965	151946	151927
T_Z	(K)	990.012	990.012	990.012	990.012	990.012	990.012
P_E	(Pa)	151680	151662	151644	151626	151609	151591
T_E	(K)	989.510	989.514	989.518	989.522	989.526	989.530
P_{SC}	(Pa)	150000	150000	150000	150000	150000	150000
T_{SC}	(K)	365.150	365.150	365.150	365.150	365.150	365.150

The "optimum" RICIN value is not known, but the different H values, when evaluated for the different RICIN values, define an H range where the best estimate of the conversion degree should fall. The overall conversion degree should be included in the range 0.5 - 0.6 and the overall burning rate should be 0.085 - 0.102 mol/s, higher than 0.068 mol/s, the maximum experimentally found value which corresponds to a hydrogen-air mixture with 5% fuel [15].

8.0 STABILITY ANALYSIS

Consider a reactor with known inlet conditions.

For an assumed value of the overall conversion degree, H', and for $0 \leq RICIN < 1$, TREE calculates the AKD' values by utilizing the balance equations of a plug-flow reactor. AKD'_{pf} is the value for RICIN = 0 and $AKD'_{lm} \doteq \lim_{RICIN \to 1} AKD'$ is the value for a stirred reactor.

TREE also calculates AKD'_{sr} for a stable operation at RICIN = 1 by utilizing the balance equations of a stirred reactor.

AKD'_{lm} may be equal to or less than AKD'_{sr}. If AKD'_{lm} is less than AKD'_{sr}, a stable stirred reactor must have a H" > H' conversion degree for $AKD = AKD'_{lm}$. Therefore, AKD'_{lm} identifies an unstable operation of the stirred reactor for H = H' (Chap. 4.1).

We will show that a "recycle" plug-flow reactor may only be unstable for H = H' if $AKD'_{lm} < AKD'_{sr}$.

1) Let us suppose $AKD'_{lm} < AKD'_{sr}$.
   We can find a conversion degree H^* so that $H' < H^* < H"$ and $AKD^*_{lm} = AKD^*_{sr}$. The relative AKD^*_{sr} and AKD^*_{pf} values will be: $AKD^*_{sr} > AKD'_{lm}$ because $H^* < H"$, and $AKD^*_{pf} < AKD'_{pf}$ because $H^* > H'$. Therefore, the AKD' vs. RICIN curve calculated for H' intersects the similar curve calculated for H^*. The intersection point identifies a recycle plug-flow reactor that satisfies two different conversion degrees. The lower value, H', corresponds to an unstable operation. The reasoning may be repeated for several H^* values and recycle plug flow reactors.

2) Let us now suppose $AKD'_{lm} = AKD'_{sr}$.
   The AKD' vs. RICIN curve does not intersect other curves which, for different assumed conversion degrees, identify stable solutions for RICIN = 1, because for each RICIN value, lower AKD values must correspond to higher conversion degrees. Every point of the AKD' vs. RICIN curve corresponds to a stable steady operation of a recycle plug-flow reactor.

For the examined mixture, we showed that $AKD'_{lm} = AKD'_{sr}$ only for $H' > 0.9$ [15]. A recycle plug-flow reactor with a conversion degree in the range 0.5 - 0.6 might therefore be unstable.

But,
- only the steady-state operation of the real reactor can be simulated by a plug-flow with a fictitious recirculation loop;
- dynamic behaviour of the real reactor could be better schematized by (2n + 1) plug-flow reactors with the fictitious recirculation loops which are shown in Figure 5;
- there is a real recirculation loop in the combustion chamber from section 2 to section 1, through F and G (Fig. 2);
- the recombiner control system regulates the gas feed flow and T_U.

For all these reasons, the instability of the recycle plug-flow reactor, which simulates the steady-state behaviour of the real reactor, does not necessarily involve a negative judgement about the dynamic stability of the real recombiner, but it does suggest the need for an experimental verification.

9.0 CONCLUSIONS

The combustion chamber of a hydrogen-oxygen thermal recombiner was originally designed to process the atmosphere of the safety containment of a water-cooled nuclear reactor in a design basis accident. The computer code TREE simulates the steady state operation of the combustion chamber and is based on an analysis of its physical and chemical phenomena. The volume where the controlled combustion takes place has been schematized with a plug-flow reactor and a fictitious recirculation loop. Two parameters in the code depend on the mixing between unburnt and burnt gases in the reaction volume. The dependence of these two parameters on the feed conditions must be deduced through experimental tests.

Since the information obtained from the tests is currently very limited, at present the code may be used to estimate working situations that differ from the experimental ones only in terms of chemical composition of the inlet mixture.

The code results show that burning rates higher than those from standard tests are also possible when nonflammable mixtures contain hydrogen and steam concentrations higher than the recombiner test values.

Under these working conditions, a "recycle" plug-flow reactor might be unstable. So, although the instability of a model which simulates only the steady state operation does not necessarily involve the instability of the real recombiner, an experimental research program should be carried out to verify whether doubts concerning instability are groundless and whether the theoretically estimated burning rates are correct. Pressure,

temperature and concentration should be measured inside the combustion chamber. TREE will be useful in programming test conditions and explaining results. The new tests, in turn, will help improve the simulation code and enable it to estimate the recombiner performance under any operative condition.

Until the results of this research program become available, an external recirculation loop could be added to the combustion chamber in order to control the inlet gas composition during accidents more severe than a design basis accident.

SYMBOLS

AKD parameter for the radical wall destruction velocity.
C_p molar heat capacity of the gas at constant pressure.
H overall conversion degree in the combustion chamber: molar ratio between the deficiency reactant burnt in the time unit and the inlet flow rate of the deficiency reactant.
J molar flow rate of the gas.
k reaction velocity constant.
k_d constant of wall destruction velocity for the unstable chemical species.

$K_j = \frac{AKD_{F1}}{AKD_j}$.

M any atom or molecule that does not take part in the chemical reaction directly but makes the product formation possible by removing the excess energy.
P pressure.
r_f molar burning rate of the deficiency reactant per unit of reactor volume.
RICIN ratio between the fictitious recirculation flow rate and the flow rate at the plug-flow reactor outlet.
T temperature.
V reactor volume.
y molar fraction.

$Y_j = \left[\frac{(y_i - y_{i,})_j}{(y_i - y_{i,})_{F1}}\right]_{H_2}$.

Δh_r heat of combustion per mole of deficiency reactant.
η conversion degree in a section of the reactor.

Subscripts

AMB relative to the recombiner inlet and outlet conditions (Figs. 1 and 2).
f relative to the deficiency reactant.
F1 relative to the F1 experimental test.
i relative to the generic chemical component or to the generic reaction in the gaseous phase.
in reactor inlet.

j	relative to the generic recombiner working condition.
lm	limit.
out	reactor outlet.
pf	plug-flow reactor.
sr	stable stirred reactor.
v	virtual.
A, E, F, G, M, SC, U, V, W, Z, 1, 1', 2	relative to the homonymous section in Figure 1.

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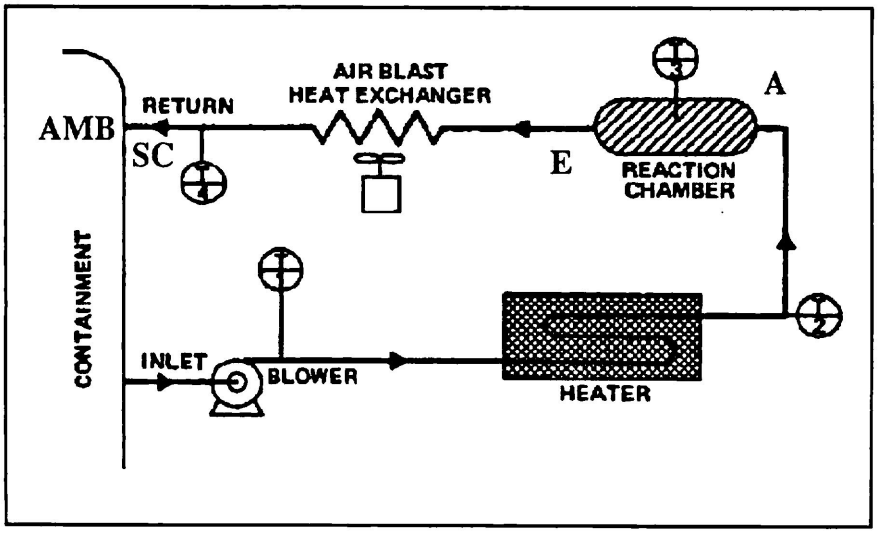

Figure 1. FIAT-CIEI Thermal Recombiner.

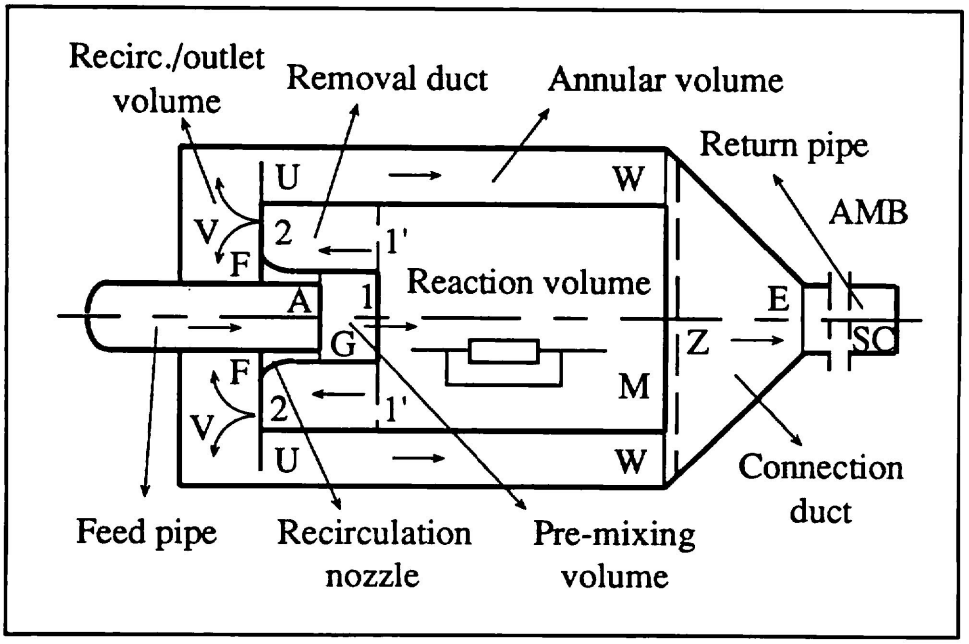

Figure 2. Combustion Chamber Scheme.

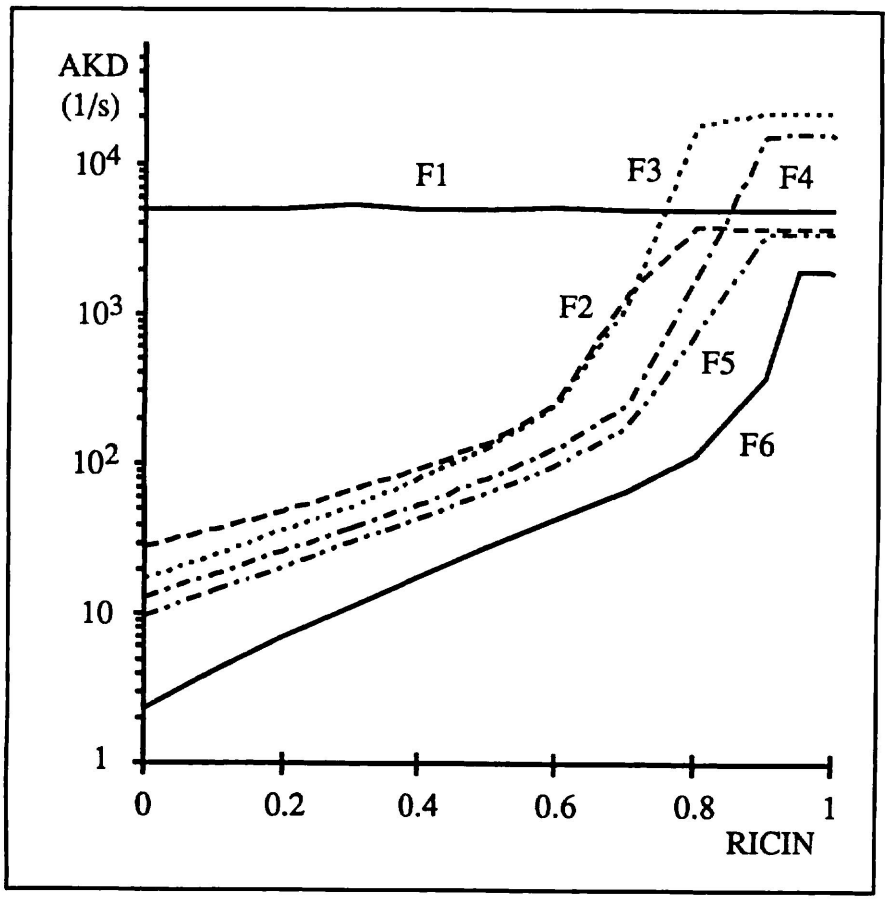

Figure 3. Pairs AKD-RICIN that Satisfy Experimental Conversion Degrees in the Code TREE.

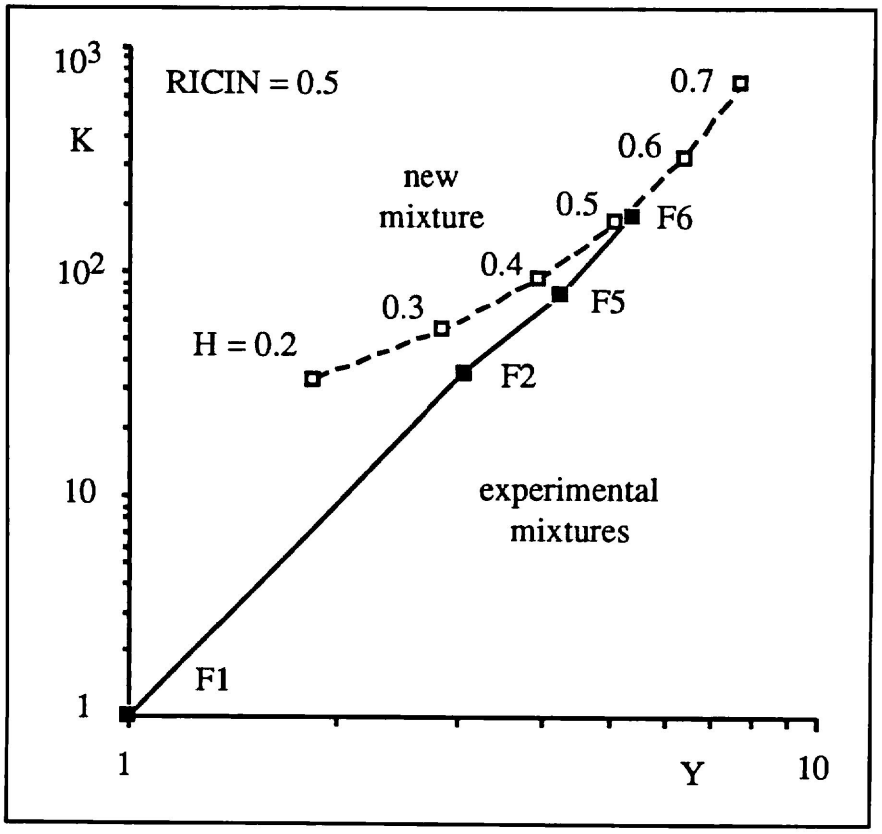

Figure 4. Graphic Evaluation of the Conversion Degree.

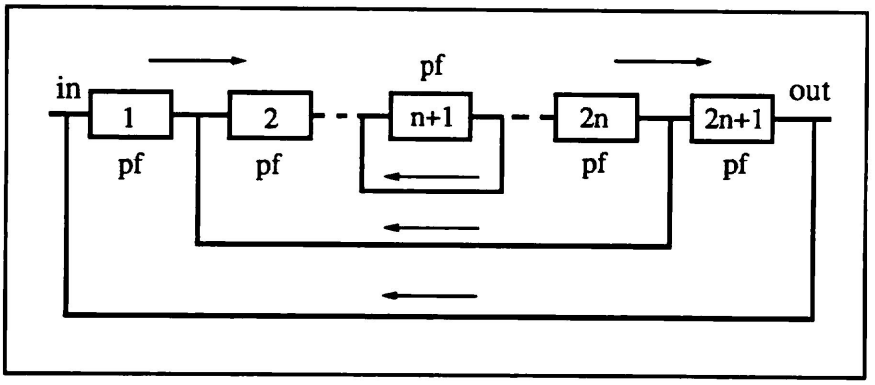

Figure 5. Dynamic Scheme of the Reactor.