

Passive Autocatalytic Recombiner (PAR) Package Reference Manual

The MELCOR ESF package models the physics for the various engineered safety features (ESFs) in a nuclear power plant. The Passive Autocatalytic Recombiner (PAR) package constitutes a subpackage within the ESF package, and calculates the hydrogen removal rate from the operation of hydrogen recombiners. This reference manual gives a description of the physical models and numerical solutions implemented in the PAR package.

User input for running MELGEN and MELCOR with the PAR package activated is described separately in the Passive Autocatalytic Recombiner section of the Users' Guide.

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1. Introduction

The MELCOR ESF package models the thermal-hydraulic behavior of various engineered safety features (ESFs) in nuclear power plants. One ESF is designed to react hydrogen in a reactor containment in a continuous manner with the goal of preventing hydrogen concentrations from increasing to levels that could produce large scale hydrogen deflagration or even detonations. There are a number of methods for achieving hydrogen removal. The most common method is the use of igniters, which provide local ignition sources that will precipitate hydrogen burns near the lower burn limits. However, these systems depend on the availability of a power source, which in certain accident sequences may be lost. In addition these systems will not operate under certain steam inerted conditions which can lead the igniter system to precipitate a large burn when inerted conditions are removed. The passive autocatalytic systems, however, do not require a power source and are not strongly affected by inerted conditions. The benefits derived from this type of hydrogen control system are obvious and are under study for possible backfitting to existing power plants.

The Passive Autocatalytic Recombiner (PAR) package constitutes a subpackage within the ESF package and calculates the rate of hydrogen removal generated by PAR type hydrogen removal systems. The default MELCOR model is based on the Fischer model [1], which is a parametric model developed for a particular type of PAR unit. The user input provides correlation coefficients for the general mathematical form of the model. These coefficients are used by the code to calculate the total gas flow rate through a PAR unit. From the PAR gas flow rate together with user provided PAR efficiencies, transient relaxation times, delay times, and the internally calculated hydrogen mole fractions, a per-PAR-unit hydrogen reaction rate is calculated. This rate is then multiplied by the current timestep and the user provided number of active PAR units to determine the change in hydrogen, oxygen, and steam masses. These differential masses are then passed to CVH as sources/sinks.

It is noted that one particular PAR design has been developed, studied, tested, and reported on in the technical literature. This type of PAR was developed by the NIS Company in Hanau, Germany. The design consists of parallel plate cartridges containing palladium-coated aluminum micropellets. It has been tested in a series of experiments as described in References [1], [2] and [3]. The Reference [3] tests are of particular interest because these tests of the NIS PAR were performed at the Sandia National Laboratories/NM *for the NRC*.

The type of PAR used as the default model in the MELCOR model is the NIS type of PAR. This type of PAR was chosen as the default model because of the literature available and because this was the type studied by the NRC; see Reference [3]. It should be noted that other PAR designs are available. However, it is likely that many other specific designs can be modeled within the parametric framework described here. In the event that it is desired

to study a design concept sufficiently different from the type described here, it was necessary to provide a more general input option. This provides the user with the option to specify a control function with which the PAR flow rate can be calculated as a function of one or more system variables. In addition, an option is provided that allows the user to specify the PAR efficiency through the use of a control function. A more detailed description of the model is provided in the next section

2. Model Description

The chemical recombination of hydrogen and oxygen to produce steam and release energy is described by the equation



The hydrogen reaction rate for a single PAR unit may be expressed in terms of the total volumetric flow rate passing through the unit as follows:

$$R_H = \eta \rho_H Q f(t) \quad (2.2)$$

where

R_H = hydrogen reaction rate (kg/sec)

ρ_H = hydrogen density of entering gas (kg/m³)

η = hydrogen reaction efficiency (~0.85)

Q = total gas-phase volumetric flow rate through the unit (m³/sec)

$f(t)$ = $\left[1 - e^{-\left[\frac{t-t_0}{\tau}\right]} \right]$ = relaxation time function during the initial PAR heat-up

τ = characteristic heat-up time (~1800 sec)

t_0 = time of PAR initiation (s)

t = time after PAR initiation (s)

The relaxation time function is intended to account for the observed transient interval before the PAR attains steady-state operation. It is thought that the primary transient effect

is due to the time required for the catalytic elements to come up to operating temperature. For the configuration tested in Reference [1], the relaxation time, τ , was determined to be on the order of half an hour.

The total volumetric flow rate through a PAR unit may, in the general case, be provided by the user through control functions. However, an expression for NIS type PAR units [1,4] has been found to accurately describe the flow rate, and is given by:

$$Q = a C_H^b \tag{2.3}$$

where

- C_H = hydrogen concentration (mole fraction)
- a = constant that depends on PAR unit design parameters (~0.67 kg/sec)
- b = exponent that depends on PAR unit design parameters (~0.307)

Some of the parameters in Equations (2.2) and (2.3) will be provided by the CVH package in the MELCOR code. For the NIS model, other than the current time, the hydrogen density and mole fraction are the only two required parameters. Depending on the specifics of the model, however, user-defined PAR flow rates may depend on other CVH parameters such as temperature or pressure.

For typical containment volumes, a number of PAR units will be required to control the H₂ concentration. The total hydrogen depletion rate is then found simply by summing the rates from the individual units in a particular control volume. The user may specify more than one type of PAR and specify the number of units of each PAR type in a single control volume or distributed in a number of control volumes.

The transient effects described by the term $f(t)$ in Equation (2.2) derives from the solution for a single step function in hydrogen concentration with the initial concentration being zero, and the hydrogen concentration in a control volume remaining constant.

In general, however, the hydrogen concentration will not remain constant and may in fact involve multiple 'bursts' of hydrogen injection into a containment volume combined with continued releases from the vessel or from ex-vessel fuel/metal, fuel/concrete interactions. It is assumed here that if there is an increase in H₂ concentration, which implies an eventual increase in volumetric flow through the PAR (Equation (2.3)), then the time dependence of the change in flow will follow the same relaxation behavior implied by Equation (2.4). Based on this assumption, a more general approach to the transient effects is employed in the MELCOR model. The transient effects are described by the differential equation:

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$$\frac{dQ}{dt} = \frac{1}{\tau} [Q_{ss} - Q] \quad (2.4)$$

where Q is the volumetric flow rate, and Q_{ss} is the steady-state flow rate implied by the hydrogen concentration found from Equation (2.3). Integrating Equation (2.4) over timestep, Δt , in which the flow rate changes from Q_{old} to Q_{new} , gives the following result:

$$Q_{new} = Q_{ss} \left[1 - e^{-\Delta t/\tau} \right] + Q_{old} e^{-\Delta t/\tau} \quad (2.5)$$

Equation (2.5) will provide for transient effects, but requires that the hydrogen reaction rate be carried as a dynamic variable with the old and new values stored in the main variable array. Note that the transient term $f(t)$ is now implicit in the flow rate equation. It should also be noted that Equation (2.5) is applied on a timestep-by-timestep basis so that the flow rates and H_2 burn rates will respond in a continuous manner to transient conditions such as increases or decreases in the hydrogen concentration. It is thus, not necessary to track the thermal response of the PAR catalytic elements. The temperature of the PAR catalytic elements is implicit in the correlations used in the Fischer model, but is not explicitly available as an output parameter.

As described by Equation (2.1), the recombination of hydrogen results in an oxygen depletion rate and a steam mass increase rate. Since the reaction is exothermic, there is an associated change in gas temperature. These rates are given as follows for the control volume in which the PAR unit is located:

$$\frac{dm(H_2)}{dt} = -R_H, \text{ kg/s} \quad (2.6)$$

$$\frac{dm(O_2)}{dt} = -\frac{M_{O_2}}{2M_{H_2}} * R_H, \text{ kg/s} \quad (2.7)$$

$$\frac{dm(H_2O)}{dt} = \frac{M_{O_2}}{2M_{H_2}} * R_H, \text{ kg/s} \quad (2.8)$$

$$\frac{dH}{dt} = \sum_{i=1}^N w_{i,in} h_{i,in} - \sum_{i=1}^N w_{i,out} h_{i,out}, \text{ W} \quad (2.9)$$

Note that in Equations (2.6) through (2.8), M refers to the molecular weight of the species. The indices on the sums in Equation (2.9), for the change in total enthalpy H , refer to the

specific gas constituents (H₂, O₂, H₂O, CO, CO₂, etc.), while w and h refer to the mass flow rate and specific enthalpy of each constituent. Also, because MELCOR uses a consistent reference point (JANIF Convention) for all gas-phase thermodynamic properties, the heating rate given by Equation (2.9) is not needed as a source in the CVH package. JANAF refers to a set of thermochemical tables [5]. The JANAF convention implicitly includes all heats of formation in the enthalpy functions for each material. In so doing, the heat of reaction, for example in the burning of hydrogen and oxygen, is included in the enthalpy of the reaction product (steam in this example). The advantage is that all chemical reactions, such as those generated in this PAR Package, can be treated simply as changes in the masses of the reactants and products, and the heat effects are accounted for automatically through the equations of state.

The mass rates computed by the PAR Package in Equations (2.6) through (2.8) will be multiplied by the current timestep and the differential masses passed to the CVH package.

The PAR testing discussed in Reference [3] did not identify any problem regarding “remaining capacity” in terms of possible degradation of the catalytic elements.

In addition, it is important to note that the literature [2] does address the investigations into possible decrease of the PAR performance because of catalyst inhibitors and poisons. The investigations performed indicate that the effects of catalyst inhibitors and poisons are negligible. However, if further studies provide evidence that catalytic elements can be degraded, an option is provided that allows the user to specify the PAR efficiency, η , by a control function that can be a function of time, aerosol concentration, etc.

The change of gas temperature as it passes through the PAR can be estimated by noting that, since no mass or energy has been added from external sources to the gas stream (with the exception noted later), the enthalpy flow rates in and out of the PAR unit must be equal. Thus, from Equation (2.10), the following relationship between the inlet and outlet conditions must exist,

$$\sum_{i=1}^N w_{i,in} h_{i,in} = \sum_{i=1}^N w_{i,out} h_{i,out} \quad (2.10)$$

$w_{i,out}$ = the mass flow rate of the i th gas-phase species exiting the PAR (kg/s),

$w_{i,in}$ = the mass flow rate of the i th gas-phase species entering the PAR (kg/s),

$h_{i,out}$ = the specific enthalpy of the i th gas-phase species exiting the PAR (J/kg),
and

$h_{i,in}$ = the specific enthalpy of the i th gas-phase species entering the PAR (J/kg).

The outlet temperature is then evaluated by a Newton's method iteration in which the inlet enthalpy flow (the left side of Equation (2.10)) is evaluated and successive estimates of the outlet temperature are calculated until the difference between the inlet enthalpy rate and the outlet enthalpy rate are effectively zero to within a specified limit.

It should be noted that the estimate of outlet temperature will not be accurate during rapid transient situations. During periods in which the PAR elements are heating up or cooling down some fraction of the energy is transferred to or from the PAR elements. In the former case the outlet temperature is over-predicted and in the latter under-predicted. Because the outlet temperature is only an output variable and does not affect any other calculations in the model or in the code, this is not a serious deficiency.

3. Discussion and Development Plans

Although the proposed model is rather simple and was developed in the form of a correlation for a specific design configuration, it is thought that it will provide the capability to accurately model the operation of the NIS type PAR unit in particular. It should also provide the capability to treat a wide variety of similar catalytic reactors given the required performance characteristics. In addition, the options that provide for a user-specified flow rate and efficiency through the use of Control Functions will provide the required additional flexibility and utility to model any type of PAR unit.

For lack of sufficient data regarding other catalytically induced reactions, the current model does not provide for the reaction of CO or other combustible species. Future improvements to the PAR models may consider these reactions.

4. References

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3. T. K. Blanchat and A. C. Malliakos (NRC/RES), "Passive Autocatalytic Recombiner (PAR) Tests at the SNL," CSARP Meeting (May 1998).
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