TOWARDS UNIFIED PROTOCOL FOR PAR'S PERFORMANCE RATING AND SAFETY MARGINS ASSESSMENT: PAR LIFE-CYCLE SYSTEMIC MODEL

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ABSTRACT

Passive Autocatalytic Recombiners (PAR) is one of the important technical mitigation means for hydrogen combustion in the NPP containments under accident conditions. For the PWR/VVER/CANDU units the PARs execute functions important for safety - reduce the local hydrogen concentration to an acceptable level and provide the homogenization of gas composition and of temperature fields in the containment. Certification and licensing of PAR technology have been accepted for the different NPP types and in the different countries on the case-by-case basement. But a comprehensive and generally accepted terminology and procedures for PAR characterization and its performance and safety rating are still absent. As a next step in PAR's technology improvement and maturity, it would be logical a development of their unified technical standardization and certification. Report is aimed to – 2) justify need in standardization of the PARs in the nuclear industry and in the hydrogen energy applications, 2) define a minimal set of the notions, which can be used for quantitative characterization of the of PARs throughout its life-cycle, 3) formulate a systemic (generic state-machine or automata) model of PAR's states under the normal and accident conditions. After verification and validation of proposed PAR systemic model it can be used as one of the starting points for the development of an international standard for PAR performance and safety.

1.0 INTRODUCTION

In accordance with the concept of hydrogen explosion safety and the requirements in the regulatory documents [1-6], to protect containment of reactor units from damage as a result of ignition (explosion) of hydrogen, which can enter the containment during accidents, on nuclear power plants (NPP) it should be provided the hydrogen accident removal system.

One of the most important technical means of mitigating the consequences of hydrogen combustion in the containment of modern NPPs in the event of accidents (design basis or beyond design basis) is the hydrogen removal system, which uses equipment whose operating principle is based on the autocatalytic recombination of hydrogen with oxygen contained in the containment atmosphere. The catalytic hydrogen recombination process can be realized by means of the passive autocatalytic hydrogen recombiners (PARs).

The objectives of this report are 1) to develop approach (systemic model) to an establishment of the uniform requirements for the standardization of PARs in the nuclear industry and the hydrogen energy applications, 2) to determine a minimum hierarchical set of concepts that could be used to quantify PAR throughout its entire life cycle.

2.0 THE NEED FOR PARS PERFORMANCE STANDARDIZATION IN THE NUCLEAR INDUSTRY AND IN THE HYDROGEN ENERGY APPLICATIONS

PARs are designed to ensure the safety of NPP units by preventing the formation of explosive mixtures in the accident localization areas and to keep radioactive substances within the established boundaries [5-12] by:

- local reduction of hydrogen concentrations in the mixture below the safety criteria to protect the elements of localizing safety systems, which ensures the preservation of the tightness and

strength of the containment and the operability of other localizing systems in normal operation and emergency situations;
- homogenization of the gas composition and temperature fields in the containment atmosphere.

The heterogeneous nature of hydrogen oxidation in PARs has the following intrinsic disadvantages of the catalytic hydrogen combustion technology:

- low specific (per unit of the catalyst surface area) rate of hydrogen oxidation in comparison with hydrogen removal in the flame;

- the formation of "hot spots" on the catalyst surface and, as a consequence, the local initiation of combustion waves ("flames") that can propagate through the gas mixture inside and outside the PAR housing. Spontaneous formation of the "hot spots" on the catalyst surface occurs due to spatial heterogeneity in the distribution of atomic centers of catalytic activity at the interface. For avoidance of the "hot spots" formation the special technological means are required during the production and processing of macroscopic catalytic elements;

- uncontrolled "catalytic ignition", which manifests itself through two possible mechanisms:

- "internal mechanism" - the formation of a self-sustaining flame near the "hot spots" of the catalyst, its propagation inside the PAR, leaving the PAR casing and further spreading independently of the processes inside the PAR. The result of the formation of "hot spots" on the catalyst surface is the local initiation of combustion waves ("flames"), independently (without the action of the catalyst) propagating through the gas mixture inside and outside the PARs housing;
- "external mechanism" - detachment / separation of the catalytic particle from the substrate / substrate, its removal outside the PARs casing, entering the area, in which the concentration and temperature are sufficient for catalytic self-heating of an individual particle.

From the above description of the physicochemical features of PARs operation, it follows that in order to perform one of the main safety functions – to keep radioactive substances within the established boundaries and to ensure the integrity of the containment, it is necessary to take into account the dual nature of the behavior of existing PARs designs under severe accident conditions.

On the one hand, existing PAR's designs can provide a stable rate of hydrogen removal in a certain range of hydrogen concentrations (from ≈ 0.1 to ≈ 5.9–6.0 vol.% H_2) [5-10]. At the same time PARs do not use an external energy source. On the other hand, there is a risk of spontaneous ignition of the hydrogen-air gas mixtures by a localized area on the catalyst surface, in which the catalyst temperature is significantly higher than the volume average value [7].

3.0 SYSTEMS APPROACH TO DETERMINATION OF THE FUNCTIONAL STATES AND PROCESSES OF RECOMBINATORS

PARs, like any other technical systems, are characterized by the quantitative (measurable or computable) parameters – performance characteristics (operational range, hydrogen removal rate, etc.) and safety margins (catalytic ignition limits, etc.).

However, in contrast to the other safety-related systems the technical standards (as the formal documents that establish uniform engineering or technical glossary, criteria, methods, processes, and practices on quality and interoperability pf the PARs) on PAR's performance and safety are still absent today.

Development of the harmonized and generally accepted by all stakeholders (research organizations - designers - manufacturers – regulators (TSO or safety supervising bodies)) technical standards on the PARs' performance and safety can be important for the following reasons:

- direct, documented evidence of the maturity of the PARs technologies,

- communication tool for effective transfer of available scientific knowledge into engineering and maintenance practice,
- decision making tool for selection of the appropriate PARs, which are suitable specifically for available needs in hydrogen safety (protection or mitigation), and/or optimization of their economic effectiveness,
- leveraging tool for the best practices promotion across nuclear industry and in the other hydrogen-safety-related applications (for example, in hydrogen energy infrastructure),
- tool for education, information, and guidance during developments of the next generation of the inherently safe PARs,
- raising performance and safety efficiency by diffusing/transferring technologies or ensuring physical and functional compatibility,
- fostering commercial communication.

In order to develop a unified standard for evaluating the efficiency of PARs and assessing the margins of their functional stability for the computational modeling of operational technical characteristics (PAR's performance, conditions of natural convection of the vapor-gas mixture through the casing, temperature and chemical composition of the gas at the outlet, temperature of the catalytic elements) and its boundaries for safe operation (concentration limits of flameless recombination) during accidents, it is reasonable to use a systematic approach to determining the functional states and processes (modes of operation) of PARs.

4.0 PAR'S LIFE CYCLE FEATURES

4.1 Main Stages of the PAR's Life Cycle

The life cycle of PAR as a technical system includes the different stages and various states (see Fig. 1), starting from the moment the need for such a system arises and ending with its complete decommissioning [12].

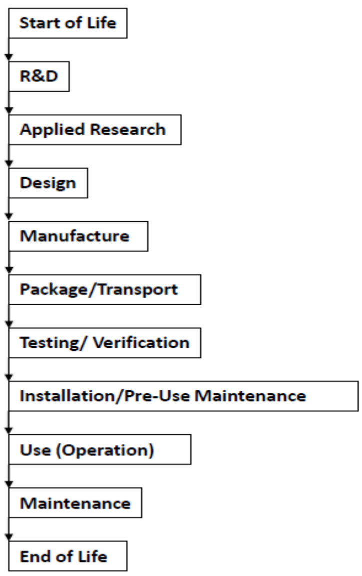

Figure 1. The main stages of the life cycle of a technical system (classical approach) [19]

In the multifactorial systems, to which the recombiners belong, at stage "Use -Operation / Functioning" various states are possible (Fig. 2). A resilient PAR can go through a series of states when it encounters a threat event that goes beyond its design constraints.

The life cycle stage features for PAR as a system are shown in Figures 2 and 3. Phase "Use (Operation /Functioning)" includes safe and unsafe (hazardous) operation. The recombiner goes into the safe functioning state from the "Ready" state. By "readiness" is meant the PAR state in which flameless catalytic recombination of hydrogen cannot be performed due to external circumstances (there are no emergency conditions, i.e. the hydrogen concentration in the atmosphere is below the minimum critical hydrogen concentration in the mixture, in which the process of catalytic oxidation of hydrogen has a stationary self-sustaining character). This parameter is analogous to the lower concentration limit of hydrogen ignition during combustion of gas vapor-hydrogen-air mixtures in three-dimensional space, i.e. fundamental characteristic of thermochemical interaction of vapor-hydrogen-air mixtures and catalyst.

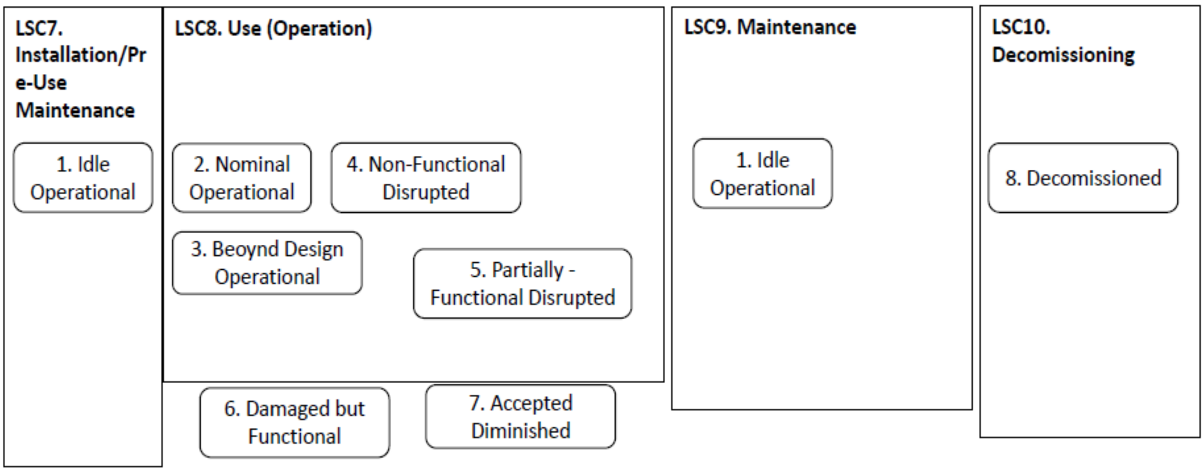

Figure 2. PAR basic states (short)

The recombiner starts up in the nominal operating state and goes into partially functional states, and sometimes completely non-functional states, depending on the event and the stability of the system (Figures 2 and 3). The processes between these states depend on the type and scenario of external events (accident scenarios), the functional stability of PARs, determined by its design, as well as decisions and actions taken by operators (aimed at preventing the development of design basis accidents into beyond design basis and at mitigating the consequences of beyond design basis accidents), and also conditions, including threshold effects (an abrupt deterioration in the level of safety caused by small changes in parameters, for example, a slight increase in the concentration of hydrogen in a mixture).

At the stage "Use (Operation)" the following main states of PARs are highlighted (Figures 2 and 3).

4.2 Nominal (Safe) Operation

The "Safe (Nominal) Operation" state means flameless catalytic recombination at the nominal capacity, i.e. the process of safe (regular) operation of PARs in the presence of hydrogen (in case of an accident). The active state of PAR, in which it has all the necessary resources and directly performs the specified functions (preventing the formation of explosive vapor-gas mixtures in the accident localization zone by maintaining the volume concentration of hydrogen in the mixture below the safety criteria to protect the elements of localizing safety systems). In this state, the recombiner is characterized by the following processes.

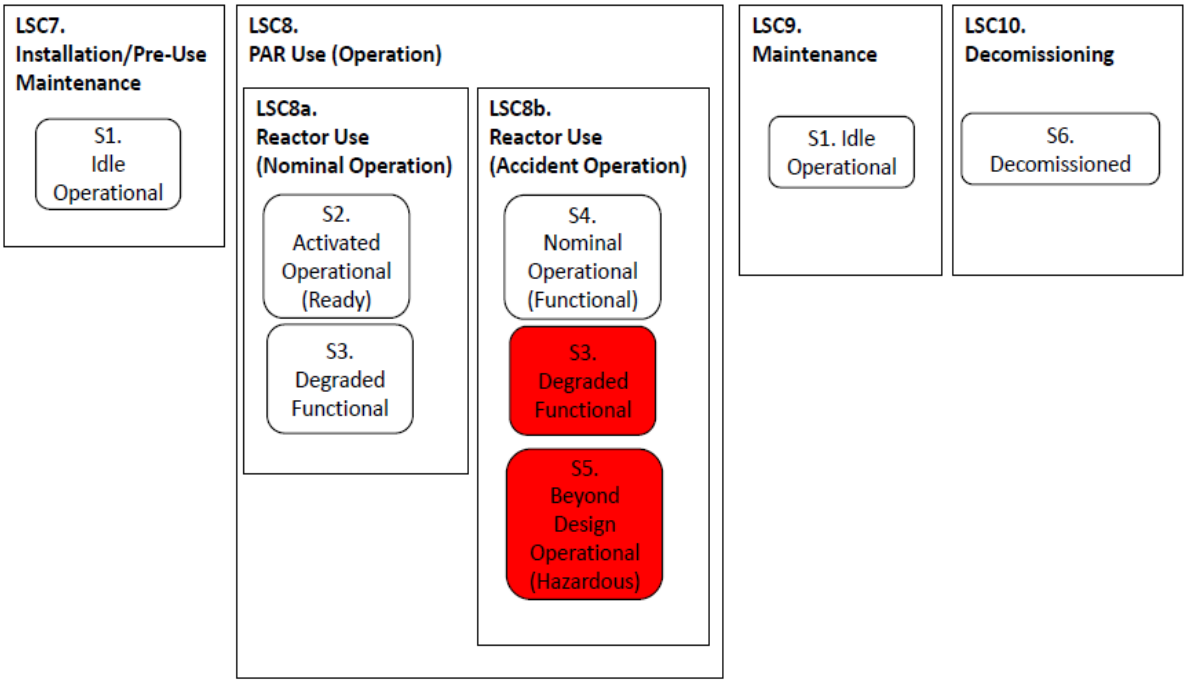

Figure 3. PAR basic states (full)

"START" = the transition process from the "READY (Idle Operation)" state to the "Safe operation" state, which can be divided into two separate sub-processes:

(a) heterogeneous ignition process, i.e. transition from the inactive state of the catalytic surface of PAR elements to low-intensity self-sustaining catalytic reactions on the surface, caused by the entry of a vapor-hydrogen-air mixture with a hydrogen concentration exceeding the lower concentration limit of catalytic oxidation at the PAR input - $c_{H_2} > c_{cat}^{crit\ 1}$;

(b) Transient process of formation of a stable, self-sustaining convection flow ("chimney effect"), i.e. the transition from the inactive state of the PAR catalytic elements to heat release due to a self-sustaining catalytic reaction on the surface, the intensity of which is sufficient for the formation and called "chimney effect"). For a fixed height of the gas-dynamic path of the PAR system and the specified geometric characteristics of the catalytic element cassettes, the "chimney" effect appears for steam-hydrogen-air mixtures with a hydrogen concentration exceeding the lower concentration limit of hydraulic start PAR $c_{H_2} > c_{chim}^{crit\ 2}$ (the power of the catalytic heat release provides a hydraulic support to the oxidation products, which exceeds the difference in the values of the hydrostatic pressure of the external atmosphere in the inlet and outlet sections).

"FLAMELESS CATALYTIC RECOMBINATION AT RATED PERFORMANCE" = the process of safe (normal) operation of PAR in an accident / hydrogen presence, characterized by:

- integral (kg / sec) and specific capacity (kg / m2sec) - PAR rate of flameless hydrogen recombination;

- integral values of the pulse of the steady-state gas jet of reagents (in the inlet section PAR) and recombination products (in the outlet section PAR);

- integral heat dissipation power.

4.3 Accident (Unsafe) Operation

"CATALYTIC IGNITION" = process of "catalytic ignition" of a hydrogen-air gas mixture when the PARS safety limits are exceeded, which can be determined by two mechanisms, internal and external.

(a) The "internal mechanism" is determined by the formation of a self-sustaining flame near the "hot spots" of the catalyst, its propagation inside and outside the PAR casing and further propagation independently of the processes inside the PAR. This process is characterized by:

- the lower concentration limit of volumetric ignition (LCLVI) of a vapor-hydrogen-air mixture caused or induced by heterogeneous catalytic oxidation of hydrogen on the surface of the catalytic elements;

- delay in time of the moment of separation from or of the flame out of the limits of the PAR protective casing after the hydrogen concentration (LCLVI) in the inlet PAR section.

(b) "external mechanism" - detachment / separation of the catalytic particle from the substrate / substrate, its removal outside the PARs casing, entering the area, in which the concentration and temperature are sufficient for catalytic self-heating of an individual particle, ignition of the gas mixture and flame propagation independent of processes in PAR. Depending on the nature (polymer, liquid, sol, etc.) and the chemical composition of the impregnation (acid, salt solution, the presence of conditioners), as well as the subsequent processing of the catalyst (drying, annealing, etc.), the structure (thickness , porosity) of the working layer itself and the value of adhesion of catalytic particles to the substrate can vary over a wide range.

Additionally, it is proposed to highlight the following PAR states.

- “NON-FUNCTIONAL DISRUPTED STATE” (Damage without preservation of functional properties). Complete inoperability and loss of all functional properties of PAR during severe accidents.

- "PARTIALLY FUNCTIONAL DISRUPTED STATE” (Damage with preservation of functional properties). In this state, although during the accident, the system suffered some physical damage, but it was able to maintain its functionality for the required period (for example, until the NPP unit returned to a controlled state, in which the fission chain reaction ceases, constant cooling of the fuel and retention of radioactive substances within the established limits). This can happen when there is sufficient redundancy in the system to maintain functional stability. In this state, the system may have a limited ability to cope with any further threats, dangerous influences. In this state, the functionality of the system has decreased, and PAR cannot be returned to its nominal state (it does not meet the established mandatory requirements - technical parameters, characteristics and functional properties specified in the technical specifications). This can happen, for example, when carbon monoxide is released during the off-frame stage of a severe beyond design basis accident and part of the catalyst surface is “poisoned” due to the presence of carbon monoxide in the mixture. However, the PAR will continue to operate at a reduced capacity and will be capable of protecting and maintaining the containment of the reactor plant from destruction.

Since to date, there is no information in the literature on the behavior of the PPVA after the appearance of catalytic ignition in it or in neighboring PARs, the self-acceleration of flames and the transition to detonation-like modes, a clear classification of the above states seems to be problematic.

5.0 CONCLUSIONS

The needs in a systemic approach to the determination of the functional states and the processes (operating modes) of passive catalytic hydrogen recombiners has been described.

Gaps in the understanding of the technical parameters, characteristics and functional properties of passive catalytic recombiners and the limits of their functional stability are highlighted and briefly described.

A systemic model (generic state-machine or automata)) of the possible states of PAR and processes is proposed, which makes it possible to substantiate a ranked list of physicochemical phenomena in normal and emergency modes of its operation when developing a metamodel of PARs behavior. Once the proposed PARs system model has been verified and validated, it could be used as one of the starting points for the development of an international standard for PAR performance and safety.

The proposed systemic model is necessary to standardize the requirements for emergency hydrogen removal systems based on passive catalytic recombiners in the nuclear industry and hydrogen energy and to develop techniques for experimental testing of recombiners, as well as to optimize and refine their numerical models.

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