WATER COOLANT TECHNOLOGY AND CHEMISTRY FOR SUPERCRITICAL WATER COOLED REACTORS

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Abstract

SCWR water chemistry technology is a big challenge for chemistry and material research. The main goal is to minimize corrosion and activity transport in the system and to ensure high purity of coolant – from active core to turbine region. So far experience from PWR and BWR reactors is used as a base for discussions of possible scenarios and options. Critical conditions could be on fuel surfaces due to high temperature and high heat flux on the surface. Also the temperature range in the active core could represent complications to find an optimal chemistry conditions.

Change in the density of SCW through the critical point is accompanied by dramatic changes in chemical properties of water. These changes mean that the ionic solubility, pH, corrosion potential etc. will be distinctly different at the core inlet compared to the outlet and system is mainly influenced by in-core radiolysis. The radiolytic yields and recombination rates in SCW are currently unknown and preliminary studies suggest a markedly different behavior at SCW conditions compared to what would have been predicted from simplistic extrapolations of the behavior encountered in conventional water-cooled.

Discussion is concentrated on different types SCWR concepts and corresponding purification and technological systems ensuring high purity of coolant and minimizing corrosion product and activity transport in primary circuit.

1. Introduction

The water regime of future reactors with a supercritical water coolant presents a challenge both from the perspective of compatibility of the medium with materials in the active zone (AZ) and from the point of radiation safety during operation of future nuclear power plants.

Water chemistry must insure the integrity of materials of the primary circuit and minimize the transport of corrosion products from the circuit to the turbine in order to minimize activity build-up at the turbine. The most critical input condition for water chemistry specification are the temperatures in the primary circuit, in the case of supercritical water reactor (SCWR) the high temperature gradient in the core plays the most important role. The water chemistry must thus respect the fact that the temperatures at inlet corresponds to pressurized water, where hydrogen dosing can be used to suppress radiolysis.

2. Materials

Materials selection and testing conditions in supercritical water is based on the presumed operational conditions of the future SCWR, which in Europe is also called the High Performance Light Water Reactor (HPLWR). The cladding temperature can reach up to 650°C and the accumulated dose can be up to 60dpa (displacements per atom). For ex-core structures, experience on materials performance can be taken from the existing fossil-fired supercritical water power plants: the materials used are either ferritic/martensitic steels or austenitic stainless steels [1]. For in-core structures, some specific issues typical of the new concept of the SCWR require further investigation. These are especially: (i) dimensional stability – creep, swelling; (ii) mechanical properties – strength, ductility, creep rupture, fatigue, creep-fatigue etc.; (iii) resistance to irradiation damage – hardening and embrittlement and (iv) chemical compatibility between material, coolant and fuel – e.g. irradiation assisted stress corrosion cracking (IASCC), pellet cladding interaction (PCI) etc. [2]. The main material groups of interest for in-core structures are ferritic/martensitic steels, austenitic stainless steels, Ni-based alloys and oxide dispersion strenghted (ODS) alloys. The most difficult challenge will likely be the fuel cladding material because the Zr alloys used in present light water reactors (LWRs) cannot be used due to their low corrosion resistance at SCWR temperatures [2, 3].

Selection of suitable materials for both in- and ex-core structures with respect to the issues mentioned above is the scope of the materials group (Workpackage 4 – „Materials“) within the European project HPLWR Phase 2, where autoclave tests are ongoing with a number of materials from the four groups of candidate alloys; in addition to tests for estimating general corrosion rates, devices are set up for tests under applied static or cyclic stresses as well. The selected candidate materials for in-core structures shall then be tested under irradiation conditions – the SCWL loop in ÚJV Řež, Czech Republic, is intended for this purpose. Weight gain tests for general corrosion rate estimation are conducted at temperatures in the range from 300°C to 650°C and at a pressure of 25 MPa, with exposure times ranging from 100 to 600 hours. The selected water chemistry conditions during the tests were selected as follows: NWC (Normal Water Chemistry), which corresponds to pure water with an excess of 125 ppb oxygen and HWC (Hydrogen Water Chemistry) corresponding to an excess of 30 cc/kg H_20 of hydrogen [4].

The following conclusions can be drawn from the results obtained: conventional ferritic/martensitic steels (such as P91, P92 or 9%Cr ODS) and austenitic stainless steels (such as 316 or 1.4970 – 17%Cr 11%Ni and 15%Cr 15%Ni, respectively) show unacceptably high corrosion rates at the highest temperatures, which disqualifies them from use as cladding materials and internals for SCWR conditions. On the other hand, acceptable corrosion rate (~0.015mm/year) were measured for alloys 800H, BGA4 (high Cr, high Ni austenitics), PM2000 (20%Cr ferritic/martensitic ODS steel) and 625 (Ni-based alloy). However, alloys 800H and 625 can be excluded for in-core structures due to their high Ni content, unacceptable for neutronics due to the high neutron effective cross-section of Ni; further, Cr content higher than 14% – the case for PM2000 – is responsible for 475°C embrittlement of these steels [4].

3. Water chemistry consideration

The primary circuit of the SCWR is a single-circuit system, which is similar in concept to a boiling water reactor (BWR), except that there is no boiling inside the core. BWRs are currently operated with normal water conditions (NWC), which corresponds to pure water, or with HWC, which uses hydrogen dosing in order to suppress the concentration of oxidizing radiolysis products and the corrosion potential.

With respect to the properties of supercritical water, plausible water chemistries in a SCWR will likely be based on dosing gases such as hydrogen, oxygen, nitrogen or possibly ammonia. It is thus recommended to use NWC and possibly hydrogen water condition (HWC) (3-6 Nml H_2/kg), as the initial water chemistry for the upcoming material tests.

In the first instance, the problems originate especially from high temperature and also great difference between the input temperature and the output temperature in the active zone (AZ). That high temperature poses a threat to the integrity of materials, mainly the coating on the fuel rods, and problems with maintaining acceptable corrosion rate in the course of operation. The great difference of temperatures may present problems in establishing optimum operating conditions – in this particular case, primarily the oxygen and hydrogen concentrations. At the input of the AZ, the temperature is within the range of readings as at the current PWR type reactors but at the output, the temperature is substantially higher. PWR reactors are operated with the hydrogen chemical regime in which hydrogen addition inhibit the oxygen concentration. Oxygen is generated by disintegrating process of a radiolytic product – hydrogen peroxide. This hydrogen should also act in the case of subcooled boiling when hydrogen is being stripped away by generated bubbles with the consequence of the hydrogen concentration reduction over the surface of the fuel pins. The PWR type and water-water energetic reactor (WWER) type reactors are currently operated with the hydrogen concentration within the range of 30-60 Nml/l. It is generally accepted that the concentration necessary to suppress oxygen even in case of subcooled boiling could be lower, with upper limit of 10 Nml/l. Hence, hydrogen addition could be used at lower temperatures with SCWR type reactors. For higher temperatures, however, there are good results with an oxygen water regime for the protection of tubing surfaces and securing adequate corrosion resistance. Nevertheless, that difference constitutes a discrepancy in the water regime concept for the future SCWR type reactors and needs to be addressed. At this stage, few experimental measurements and data verifying a radiolytic model of water dissociation in the AZ are available to help with selecting the appropriate water regime.

In the second case, which is the radiation safety of a nuclear power plant with a SCWR reactor, the important criterion is the transport of radioactive corrosion products from the AZ to the turbines. The water regime concept and coolant technology must prevent the radioactive contamination of turbines and so allow free access for the maintenance and repairs of the machines. It is well documented, that the staff performing maintenance work on the steam-generators of the PWR and WWER have the highest radiation exposures. SCWR reactors are single-loop types; as a result the steam goes straight to the turbines – this is similar way to BWRs, but BWRs have circulation of coolant in the AZ by means of
re-circulating loops, which provide also coolant purification and treatment. SCWR, however, all water from the AZ comes to the turbines. Because of the above given differences, operational experiences with BWR reactors are only partially applicable.

4. Corrosion product release and transport

At ambient conditions, a thermodynamically unstable compound might form a protective layer consists of its salts – typically an oxide. The stability of this layer is a rate-determining factor of corrosion. Corrosion can proceed i) locally - Higher temperatures increase the number of local defects and also increase the tendency of the oxide film to incorporate anions. These phenomena lead to a very dangerous form of corrosion – pitting. Its appearance is stochastic and non-predictive. Another location threatened by local corrosion are grain boundaries. Intergranular corrosion caused by different chemical composition of grain boundaries can occur. Metal carbides and nitrides are formed and the inhomogeneous composition (for example enrichment or segregation) of alloy enables local electrochemical cell formation and another detrimental corrosion mechanism can be observed. The amount of dissolved material and the penetration depth are regularly low. As a result, this form of corrosion is not very dangerous. One of the most dangerous corrosion forms that can occur in SCW is stress corrosion cracking (SCC). Its occurrence is stochastic, and as a result failures can be catastrophic. SCC is observed along the grain boundaries or through the grains. The most detrimental anions causing the emergence of SCC are chloride, bromide and sulphide. SCC needs both chemical and mechanical components of stress. SCC commonly leads to a failure of the entire reactor. ii) Generally - Another form of corrosion which occurrs in supercritical water is general corrosion. General corrosion happens in case, where none of the alloy components could form a protective layer, or where the protective oxide film is unstable. It has been shown for different alloys and stainless steels that pitting switches to general corrosion above a certain temperature called “the inversion temperature”.

Stability of above-mentioned protective layer is determined by two parameters – its high kinetic stability and low dissolution rate. Kinetic stability means in this case that the layer is a barrier between the solution and metal and protects it from corrosion. A low dissolution rate means that it has a low tendency to dissolve [5].

When those conditions are not met, then the construction materials or protective oxide layer made by corrosion products can pass into solution. Conditions of corrosion product release and transport in an SCWR system are dependent on properties of fluid, properties of mass (particle) and properties of material surfaces. Processes controlling mass (particle) transport are: i) diffusion, ii) thermophoresis, iii) turbulence, iv) gravity, v) electrical forces, vi) Van der Waals forces [6].

The main process of release of material into solution is dissolution of corrosion products. The basic difference between current power plants and an SCWR is that a coolant in a primary circuit of BWRs and PWRs changes state from liquid to gas. The coolant in a primary circuit of a SCWR does undergo a transition from “water-like” to “steam-like” (from ~0.8 to 0.1 g/cm^3) densities as it passes from subcritical to supercritical conditions, through the critical temperature. Below the critical point is water able to dissolve metal ions from its oxides and hydroxides [7]. When the temperature passes the critical point the hydrogen bonds are broken, which is responsible for the decrease in the dielectric constant, changes in the dynamic viscosity and the increase in the self-diffusion coefficient of water. This will result in the change in nature of water as solvent – a decrease in dielectric constant on going through the critical point causes an increase in the solubility of organic compounds and a decrease in the solubility of inorganic ionic compounds [8]. In temperature range between 275 and 450°C, 25MPa (pseudo transition zone) very unique chemistry occurs. The implications of this chemistry for corrosion product transport have yet to be fully elucidated. Above 450°C, SCWR steam-like solid-gas models can be used [7]. The solvent ability is also dependent on water density. Subcritical water has higher density, supercritical water has lower density. The solvent characteristic is very important for understanding the process of formation of corrosion product deposits. Metal ions released in the high water-density locations, where the metal oxide solubility can be high for many metal ions, can be transported to downstream low-density regions and precipitate, resulting in reduced thermal transfer efficiency and deposit formation [9]. The corrosion mechanism in high density regions is as follows: Low conductivity caused by decrease in dielectrical constant hampers ion transfer between the centres of the conjugated redox reactions (electrochemical mechanism of corrosion is suppressed). Then is one-step reaction of metal and oxygen. Metal oxides, which form the protective oxide layer of construction material, become reactive in supercritical water conditions and dissolve [8]. The most important oxides are likely magnetite, chromic oxide and nickel oxide. The solubilities of these three oxides decrease sharply as the critical temperature is traversed, which has significant implications for deposition on the fuel and activity transport throughout the power generating circuit [10].

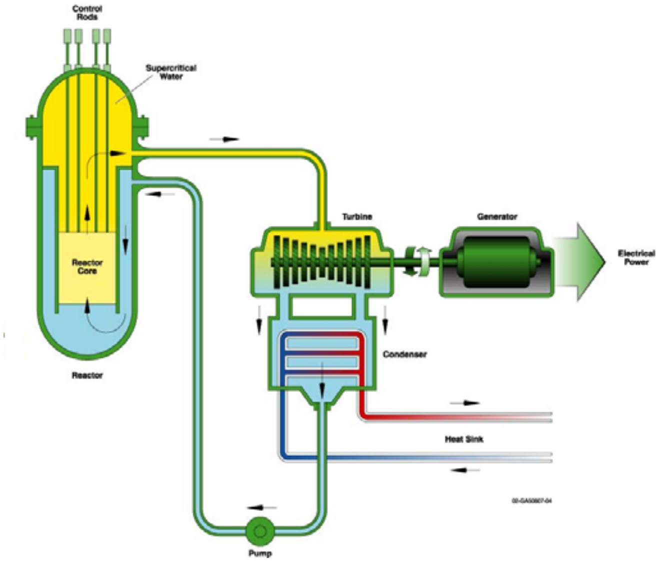

Fig. 1: Schematic design of a supercritical water-cooled reactor [11]

Fig. 1 shows diagram illustrating one schematic concept of the SCWR. The yellow part represents the low-density region and blue part the high-density region. The process of above-mentioned corrosion product release and transport is as follows: water in the core is heated to supercritical parameters, and then goes to the turbine and condenser, giving up heat, and its temperature decreases to ~280°C. It then becomes high-density gas, and in the blue part of diagram the dissolution of corrosion products and other impurities takes place (heaviest deposits were observed at temperatures around the critical point [7]). Those dissolved species will be transported in-core and precipitates on surfaces (especially surfaces of the fuel cladding), where water makes the transition into the supercritical region [12].

High levels of release and transport of corrosion products have very unpleasant consequences, because the radioactive corrosion and fission products are transported out of core, and became deposits, especially in the core. This reduces heat transfer and increases the possibility of fuel failure. The high level of corrosion product transport increases the out-of-pile radiation fields and worker dose. The coolant of the primary circuit doesn’t transport just corrosion products [7]. The transport of impurities, which can concentrate, also takes place when the water is inadequately prepared or construction materials are inappropriately chosen, e.g. copper and silica species. These species are volatile in steam and form turbine and condenser deposits. This complicates maintenance of turbine, because the deposits of the primary circuit are radioactive. It is therefore necessary to ensure sufficiently reliable and effective purification of the primary coolant [13]. Extremely pure coolant should be used at SCWR plants. Full flow condensate demineralization and pre-reactor mechanical cleanup of feedwater by means of high temperature filters should be used in SCWR (more details are given in the section “SCWR coolant purification”) [14]. To ensure, that chemical conditions meet requirements, the following control parameters should be checked: i) conductivity, ii) pH, iii) concentration of dissolved ions and redox species (especially radiolysis products H_2 and O_2), iv) particulate content [13]. These parameters can influence the proper choice of water chemistry regime. Some water chemistry regimes, typically used in fossil-fired SCW plants, are listed in Table 1. Those water treatments might be suitable for the primary circuit of a SCWR, but with radiolysis must be taken into account [7]. The proper chemical regime enables: i) reduced corrosion product transport and release, ii) reduced flow accelerated corrosion (FAC) of the feedwater system piping, iii) reduced maintance costs. Proper chemical conditions in the primary circuit ensure i) minimizing corrosion, fouling, activity and corrosion product transport and deposition, ii) optimizing thermal performance, iii) maximize component lifetime [13]. More details about water chemistry regimes will be given in the section "Specification for water chemistry limits".

Table 1: All-Volatile and Oxygen Water Treatments used in Fossil-fired Supercritical Power Plants [7]

Water chemistry	pH at 25°C	Details	Reference
Ammonia + hydrazine	-	0.7 - 1 mg amine/kg	[15]
	9.1 - 9.4	$NH_3 + N_2H_4$	[16]
	8.5 - 9.5	NH_3	[17]
	9.1 - 9.6	-	[18]
	9.1	NH_3<0.8mg/kg to avoid copper transport	[19]
	>9	NH_3 or cyclohexylamine + hydrazine	[20]
Hydrazine only	8 - 8.5	N_2H_4 at 60 - 100 µg/kg	[16]
	7.7	N_2H_4 at 60 - 100 µg/kg	[21]
Chelate + NH_3 + N_2H_4	-	80µg/kg chelate, 0.8 mg/kg ammonia, 0.2 mg/kg hydrazine	[22]
	-	Increases thermal conductivity of iron oxide deposits	[19]
pH 7 with oxygen	-	50 - 200 µg O_2/kg, conductivity <0.1 µS/cm	[15]
	6.5 - 7.3	-	[19]
	-	Reaction of $Fe(OH)_2$+O_2 slow compared to reaction with H_2O_2	[18]
	-	<0.15 µS/cm, 0.2 - 0.4 mg O_2/kg at boilet inlet	[23]
Combined mode	8 - 8.5	$NH_3 + O_2 \leftrightarrow NH_3$ provides slight buffering of pH so that impurities do not cause wide swings in the pH, especially to the acid side where corrosion rates can increase	[23]

5. Behavior of organic substances in SCWR circuits

In supercritical water very many of different types of organic reactions can take place. Ongoing reactions are very dependent on the conditions in circuit – especially pressure and temperature, because these conditions determine the state of water (especially density) which determines the dissolution characteristics of water.

In the subcritical region the dielectric constant reaches its maximum and subcritical water is a rich source of H^+ and OH^- . It can serve not only as a polar solvent, but also as a selfneutralizing acid-base catalyst for hydrolysis reaction. When the critical point is reached the dissolution characteristic is changed. Hydrogen bonds are broken and the reaction environment become non polar. Differences between the physical properties of subcritical and supercritical water are responsible for the fundamentally different mechanisms of organic reactions proceeding in these media. Water in supercritical state has in organic reaction those functions: i) solvent, ii) reagent and iii) catalyst. [8] As a result, in pure SCW very many types of organic reactions can occur, and it is not simple to predict how an organic compound will behave in the specific system. Six basic groups of reactions take place in SCW: i) cleavage/hydrolysis, ii) elimination, iii) condensation, iv) synthetic reactions, v) organometallic reactions [24]. The main parameter, determining the properties of water as an environment for chemical reactions is the content of oxidizing and reducing agents. Supercritical water can be mix of water, oxygen, hydrogen and hydrocarbons in any proportion; this fact has very intensive influence of ongoing reactions. Generally, increasing temperature (and decreasing water density) increases the yield of simple compounds (f.e. CO, CO_2), but generally the contact of supercritical water and organic compounds leads to the formation of many compounds [25]. If an oxidant (mostly O_2) is present in the medium, organic compounds react completely in a single-phase reaction forming CO_2 and H_2O (as used in supercritical water oxidation) and if heteroatoms are present, they are transformed into the mineral acids (HCl, H_2SO_4, …). These mineral acids can react with cations present and salts are formed, which can precipitate from solution and complicate the operation of the reactor [26].

6. SCWR coolant purification

At the Heidelberg symposium Kozinski [27] presented an overview of potential application of high temperature filtration and separation technologies that can be considered for purifying supercritical water. In the next part of this paper primary coolant high temperature technology used in VVER pressurized reactor is described. This is one technology for possible use in a SCWR.

A high temperature purification system (HTF) is used in VVER 1000 units and has the following main function of primary coolant impurity and radionuclide clean up using high temperature filters.

In VVER-1000 V320 units the primary circuit letdown filtration system (HTF) is located in the primary circuit (Fig.2,3). The primary circuit has four identical loops, each containing a high temperature high flow rate mechanical filter containing a titanium metal sponge filter medium (Fig.4), followed by a catcher filter. The four loops are connected across each of the main coolant pumps and the flow rates are determined by the pressure (0.62 MPa) developed across each pump. The filters operate at 290°C, 15.7 MPa and have a design flow rate of 60 to 80 tonnes/h per loop (100 tonnes/h per loop maximum, maximum 0.5% of the total coolant flow).

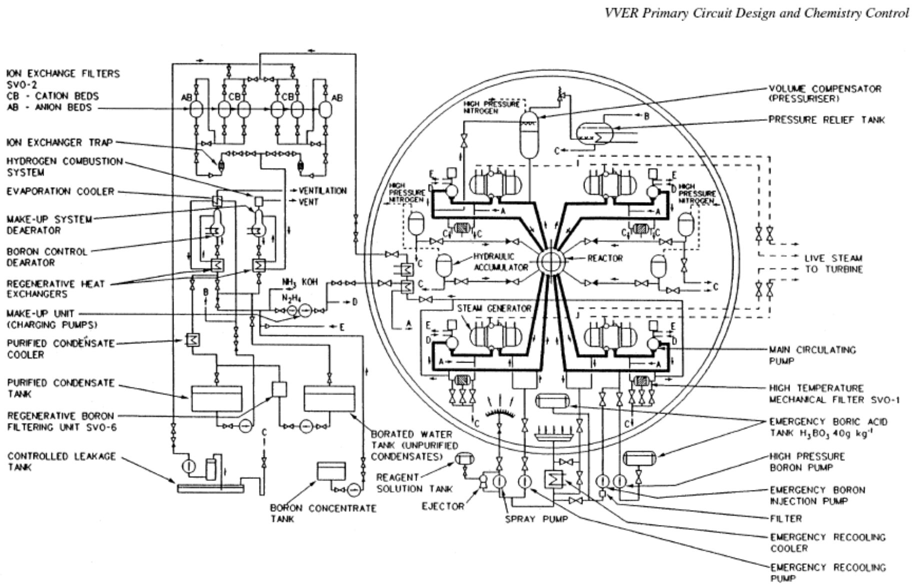

Fig. 2: Schematic diagram of the primary and auxiliary circuits of a VVER 1000

The filters are intended to remove fine particulate thus reducing radiation fields, but as the coolant should normally contain only small concentrations of particulate they are probably only effective at start-up and shutdown.

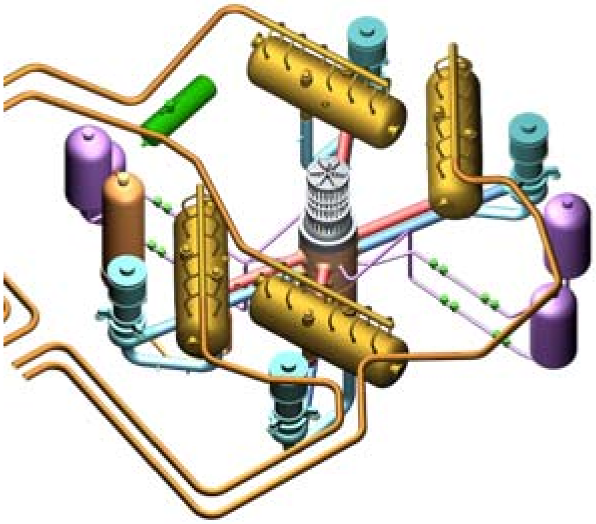

Fig. 3: VVER 1000 primary circuit with HTF filters

The main effect of the high temperature filtration is the redistribution of dissolved and dispersed particles that are present in the coolant. The contribution of dissolved activity in the coolant to the total activity in the coolant is 10x higher than the activity of dispersed particles. With HTF usage, the dispersed iron concentration in the coolant volume is 2-5 ppb, 10x lower then VVERs without an HTF. This also leads to much lower activity of Co60 and Co58 in coolant.

The full time operation of HTF systems leads to one disadvantage. During transients (startup, shutdown) the mass transfer of corrosion products takes place, and is related to the change of solubility of corrosion products that is caused by the change of thermal parameters of the primary coolant. As a result, the activity of corrosion products increases several times in the reactor coolant. This can be also one of the reasons for a little drainage effect – most of drainage possible outlet is outwash during the shutdown process due to solubility changing [28].

The filtration efficiency [29] can be described as the relation between the concentration of particles on the filter input and the input/outlet concentration ratio. At low values of inlet concentration the outlet concentration could be higher; this means that corrosion products are released from the filter (Fig.5).

Fig. 4: HTF titanium filtration material

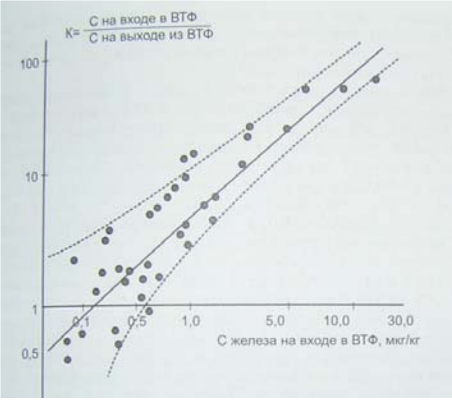

Fig. 5: The filtration efficiency described as a relation between the concentration of the particles on the filter input and the input/output concentration ratio

7. Specification for water chemistry limits

It has been mentioned that chemistry control in an SCWR is likely to be a hybrid of current practices from both the fossil and nuclear power plants [13]. In the SCWR reactor layout the feedtrain lines are the source of corrosion products entering the SCWR core. Based on BWR experience we can discuss possible limits of corrosion products mainly iron in the inlet to the core and corrosion products in core.

German and some other EU BWR units can be good example for practical use. German BWR plants differ from other plants e.g. US BWR´s in using only normal water chemistry (NWC) without any other chemistries or additives, such as hydrogen water chemistry (HWC), zinc injection, noble metal addition, etc.[30].

While the optimum chemistry is not yet specified, EU BWR water chemistry specification is good first example on SCWR chemistry specification. There are two main important parameters could be considered first, conductivity and iron content. In all BWRs in Europe the conductivity in the reactor coolant is below 0,1 micro Siemens/cm, iron content in reactor coolant is about 2 ppb. Feedwater iron concentration is different at different plants, about 0,5 ppb iron can be achieved. Oxygen in feedwater is between 50-100 ppb. Achieving 0,5 iron corresponds to the proposal made by Guzonas [13] for the SCWR core inlet to avoid in-core deposition.

8. Conclusions

Water chemistry must insure the integrity of materials of the primary circuit and minimize the transport of corrosion products from the circuit to the turbine in order to minimize activity build-up at the turbine.

The concept of the EU SCWR is based on a reactor pressure vessel similar to a pressurized water reactor (PWR), but designed for a higher pressure of around 25 MPa with an inlet temperature of 280o C and an exit temperature of 500o C.

With respect to the properties of supercritical water, water chemistry will be likely based on dosing gases such as hydrogen, oxygen, nitrogen, ammonia or hydrazine reflecting temperature gradient in core. The most important requirement is to avoid in-core deposition, which could be achieved by a combination of low iron input to the core, hot filtration and proper water chemistry.

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