STRATEGIES FOR CORROSION MONITORING AND CONTROL IN A CANDU®-SCWR

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Abstract

To minimize corrosion and corrosion product transport in a reactor, it is necessary to implement a chemistry regime, and monitor and control the chemistry to within specifications. This paper highlights three areas of concern for chemistry control in a supercritical water-cooled reactor (SCWR), reviews water chemistries used in fossil-fired and nuclear power plants, discusses modern techniques for high-temperature chemistry control and monitoring, and discusses how chemistry control might affect design. The paper concludes by summarizing the challenges for chemistry control and monitoring in a SCWR, and identifies the need for chemistry control and system design to work together.

1. Introduction

The long-term viability of any Generation IV SCWR concept depends on the ability of reactor developers to predict and control water chemistry in order to minimize corrosion and radionuclide transport associated with the new design. While some recent proposals have considered indirect-cycle designs, this paper will only consider a direct cycle design with a core inlet temperature around 350ºC (i.e., subcritical temperatures) and a core outlet temperature around 625ºC (Figure 1). The chemistry requirements for this design include:

- control of water chemistry to minimize corrosion of the feedwater system;
- minimization of corrosion product transport, in-core deposition and radionuclide transport through choice of water chemistry and/or purification;
- control of water radiolysis in order to minimize corrosion and stress corrosion cracking of in-core and downstream components;

There is a strong dependence of materials performance on coolant chemistry in all nuclear power plant systems. To achieve the design life of reactor components it is necessary to monitor and control relevant chemistry parameters, such as conductivity, pH, concentrations of dissolved ions, concentrations of redox species (e.g., H_2, O_2) and particulate content. Chemistry performance requirements are set by the sometimes conflicting desires to minimize corrosion (general and localized), fouling and activity transport, optimize thermal performance and maximize component lifetime. The primary requirement of chemistry control is to reduce material degradation rates such that design lifetimes are achievable for the entire system. The complexity of the systems, the need to use diverse alloys (e.g., steels and Zr alloys), reactivity requirements and limitations on chemical additives mean that coolant chemistry is always a careful balance. Typically, chemistry control is achieved by system

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design, the use of chemical additives, and by operational methods (e.g., purification). A key requirement of any chemistry control regime is that chemistry "control parameters" must be monitored and adjusted within the specified timeframe.

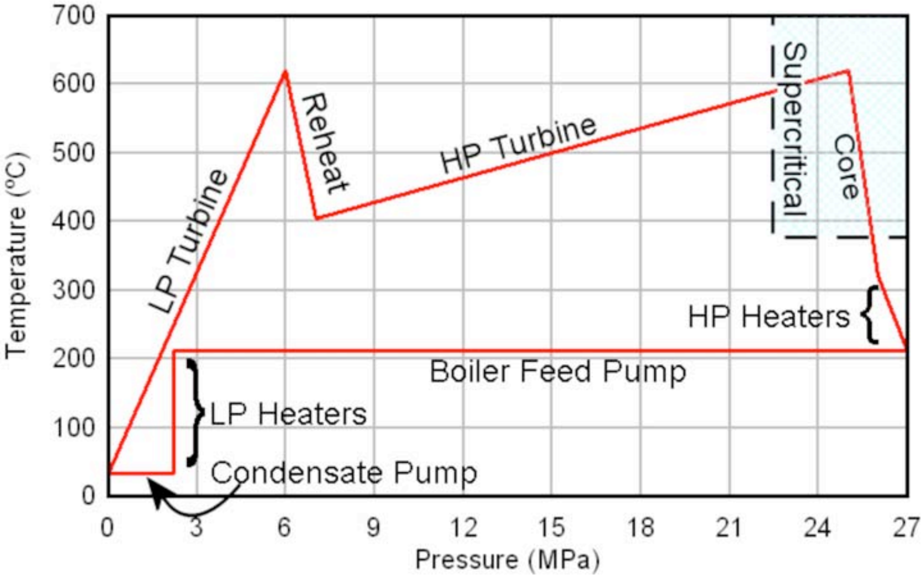

Figure 1 Temperature-pressure diagram of a representative supercritical Rankine cycle. Changes in temperature and pressure resulting from the operation of one or more components are indicated.

Typical chemistry control regimes in fossil-fired supercritical water plants have evolved from industry practices, and have been designed to reduce corrosion product transport, flow accelerated corrosion (FAC) of the feedwater system piping and maintenance costs. These chemistry regimes are a good starting point for a direct-cycle SCWR; however, in a nuclear power plant, the major focus must be on minimizing the possibility of core failure, whether through degradation of reactor internals, fuel fouling, or degradation of coolant piping, and therefore chemistry strategies that work well for a fossil-fired SCW plant may not be adequate for a SCWR. An additional consideration is that chemistry control options for nuclear plants are limited due to the effects of radiation on potential chemistry control additives. Therefore, chemistry control in a SCWR is likely to be a hybrid of current practices from both the fossil and nuclear industries.

Existing methods of chemistry monitoring in nuclear power plants are predominantly ex-situ (cooled and de-pressurized) and off-line (batch laboratory analysis of grab samples). The adequacy of this strategy varies with the sampling location and the chemical parameter being monitored. For example, dissolved oxygen is known to be consumed by reactions on the surfaces of the tubing of the sampling systems. These issues will become even more serious for chemistry monitoring in a SCWR as a result of the large changes in water chemistry around the critical point. In addition, grab sampling in nuclear systems can be labour and dose intensive. It is likely that, for key chemical parameters, reliable monitoring can only be achieved through the development of in-situ or on-line probes. In addition to their ultimate use in a SCWR, these monitoring techniques will be needed in the short term for use in any in-reactor test facility.

The objective of this paper is to describe the key chemistry control issues expected in a SCWR, and to propose possible monitoring and control strategies to address these. The implications of these strategies for both materials selection and design are highlighted.

2. Chemistry control issues in a CANDU-SCWR

There are three key areas of concern for chemistry control for a CANDU-SCWR design:

1. Degradation of core internals (fuel cladding, ceramic insulator, metallic liner);
2. Degradation of downstream piping;
3. Corrosion of feedtrain piping and corrosion product transport.

A brief discussion of each of these areas of concern follows.

2.1 Degradation of Core Internals

Figure 2 illustrates one concept for a CANDU-SCWR fuel channel design. The fuel channels will experience water temperatures from 320ºC at the inlet to as high as 620ºC at the outlet, with the peak fuel cladding temperatures as high as 850ºC [1]. Additionally, fuel channel materials will experience the effects of radiation, including both direct (by interaction of radiation with the alloy) and indirect effects (by interaction of radiation with water and subsequent interaction of radiolysis products with the alloy surface). As such, materials with good corrosion resistance and irradiation properties will be needed. Although there is significant industry experience with the use of SCW in fossil-fired power generation, there are significant differences in both structural design (thick-walled boiler tubes vs. thin-walled fuel sheathing) and material requirements (boiler tubes have a service life exceeding 20 years [2], whereas SCWR fuel sheathing has a service life of 6 years) that limit the applicability of this knowledge to the SCWR.

The liner material and ceramic insulator are unique to the pressure tube design, although the liner material can benefit from research performed in support of the pressure vessel design. A key concern is the effect of water radiolysis products on the porous insulator material, and the possibility of galvanic coupling between the liner alloy and the pressure tube, mediated by the (potentially) oxygen-conducting ceramic.

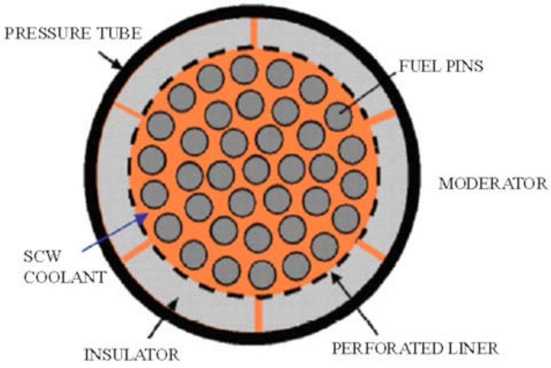

Figure 2 An insulated pressure tube design for the CANDU-SCWR.

2.2 Degradation of Downstream Piping

The second area of concern is the piping immediately downstream of the reactor core. The CANDU-SCWR concept uses an array of feeder pipes (or possibly channels) to collect the water from the individual fuel channels and combine it into common piping. With the exception of the in-core components, this piping will experience the most aggressive temperature and chemistry conditions. Recent experiences with FAC and cracking of the carbon steel outlet feeder pipes at CANDU plants [3] have highlighted the importance of materials selection and optimum chemistry control to minimize degradation of these components. While FAC may not be an issue for these components in a SCWR, other modes of degradation (general corrosion, stress corrosion cracking) may be important.

It has yet to be determined if the addition of hydrogen (or some other additive) can be used to suppress the net radiolytic production of oxidizing species [4]. If the formation of oxidizing species cannot be controlled, the coolant immediately downstream of the core could be highly oxidizing (oxygen is completely miscible in SCW); some means of measuring the concentrations of the oxidizing species and determining their lifetime in the piping downstream of the reactor core will likely be required.

2.3 Corrosion of Feedtrain Piping and Corrosion Product Transport

The third area of concern is the feedtrain piping, the source of all corrosion products entering the SCWR core[1]. The release and transport of corrosion products from the surfaces of system components has been a serious concern for all water-cooled nuclear power plants. The consequences of high levels of corrosion product transport are:

- increased corrosion product deposition on fuel cladding surfaces, leading to reduced heat transfer and the possibility of fuel failures, and

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[1] It is expected that full-flow condensate polishers would be implemented at the condensate pump outlet to minimize the ingress of corrosion products into the feedtrain.

- increased production of radioactive species by neutron activation, resulting in increased out-of-core radiation fields and worker dose.

Very little is known about how corrosion products may behave in a SCWR, but examination of data from fossil-fired SCW power plants suggests a significant risk of deposition of corrosion products released from out-of-core surfaces onto fuel cladding surfaces in-core, even when materials with low general corrosion rates are used in the feedtrain piping. The deposition occurs during the transition from liquid-like to gas-like fluid through the pseudocritical temperature, where the properties of SCW change dramatically (Figure 3).

Using a simple model of Fe transport in a SCWR and input parameters extrapolated from subcritical temperatures, Burrill [5] predicted that for Fe-saturated coolant at the channel inlet, a peak deposit weight of 108 mg Fe/cm^2 could be obtained. This is about 10 times that seen in Russian fossil-fired SCW plants, and 10^4 times higher than values typically found on CANDU reactor fuel sheaths. It has been recognized that the current CANDU reactor iron transport model over-predicts the measured in-core deposition by factor of 100 [5]; the error is believed to be a result of the effects of radiation on solubility, which have yet to be modelled. However, even assuming that such a factor can be applied in SCW, the amount of in-core deposition could still be 100 times higher than in current CANDU reactors, leading to unacceptable consequences. Therefore, some attention must be paid to minimizing corrosion and metal release from components in the feedwater system.

Corrosion product transport from the reactor core is also a serious concern, as these corrosion products can be radioactive. The turbines of nuclear and thermal power stations all experience deposition of copper and silicon species, which can be volatile in steam, at a level that may cause turbine failure. Experience at fossil-fired SCW plants suggests turbine deposits will be higher in a SCWR. A more serious problem will arise if activated corrosion products, fission products or actinides are soluble in SCW. Boiling Water Reactors (BWRs) have experienced vapour-phase transport and turbine deposits of ^{16}N, ^{58}Co and ^{60}Co when their coolant chemistry was changed from oxidizing conditions, in which nitrogen exists as non-volatile NO_3^-(aq), to reducing conditions with NH_3(aq). While a BWR core contains distinct water and steam phases, so that non-volatile species are trapped in the water, the coolant of a SCWR will transport all aqueous species along the core at progressively increasing temperatures until they react to deposit as precipitates or form volatile species in the "steam-like" supercritical fluid. Activity transport is therefore likely to be more severe in a SCWR than in a BWR, and to involve more species.

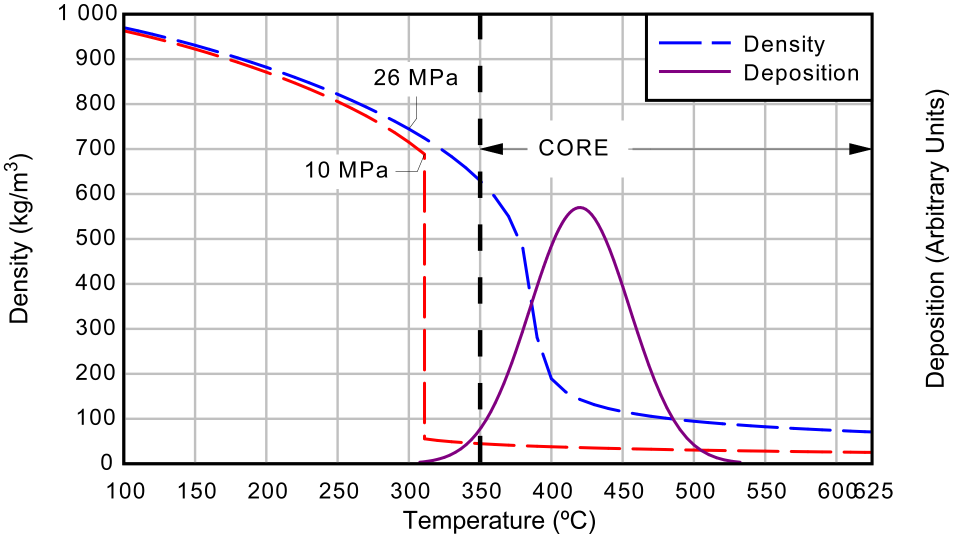

Figure 3 Potential corrosion product deposition in a direct-cycle CANDU-SCWR. The dashed lines show the density at 10 MPa (current reactor operation) and 26 MPa (SCWR operation). The solid line shows the predicted corrosion product deposition profile in SCW, based on fossil-fired SCW plant experience.

3. Water Chemistry, Monitoring and Control

3.1 Water Chemistries

Several water chemistry regimes are typically used in fossil-fired SCW plants [6]: ammonia-hydrazine, hydrazine only, and oxygenated treatment (OT) (neutral pH plus added oxygen, and the combined treatment, consisting of injection of oxygen or hydrogen peroxide into the feed water to give 50-300 µg/kg dissolved oxygen plus addition of ammonia to give a moderately alkaline pH between 8.0-8.5 at 25ºC). Table 1 summarizes some of the details on water chemistries used in supercritical power plants. No SCW plants use a non-volatile alkali such as KOH or LiOH for pH control, as is the practice in PWRs and Pressurized Heavy Water Reactors (PHWR). Recent work [7] has shown that LiOH may be viable as a pH control agent in SCW, as it is soluble under supercritical conditions, but its propensity to deposit on turbine blades will have to be determined.

There is some relevant experience in chemistry control in reheated nuclear steam in Russia based on the operating experience of the pressure-tube boiling water reactor AMB-2 of the Beloyarsk Nuclear Power Plant [8], which used nuclear steam reheated to 510ºC. The first chemistry control measure implemented was the addition of hydrazine-hydrate to the feedwater and condensate of the high-pressure feedwater heaters, in order to remove radiolytically-generated oxygen carried in the steam (>5 mg/kg O_2), and a decrease in corrosion product transport was reported. Later, ammonia was added in concentrations up to 1 mg/kg to control pH and to suppress the net radiolytic production of oxygen. The combination of ammonia and hydrazine-hydrate addition to the feedwater was reported to lower oxygen concentrations to 0.01 mg/kg and iron oxide concentrations to 0.03 mg/kg.

However, recent work**Error! Reference source not found.** [10] has found high metal release in SCW solutions containing ammonia or ammonia and hydrazine, probably as the result of the formation of metal-ammonia complexes. Further studies of the effects of water chemistry on radiolysis and corrosion product transport are required.

Table 1
Survey of Water Chemistries Used in Supercritical Fossil-Fired Power Plants

Water Chemistry	pH at 25ºC	Details	Reference
Ammonia + Hydrazine	8.5 - 9.6	0.7-1 mg amine/kg $NH_3 + N_2H_4$	[12]
Hydrazine Only	7.7 - 8.5	N_2H_4 at 60-100 μg/kg	[13]
Chelate + $NH_3 + N_2H_4$	-	80μg/kg chelate, 0.8 mg/kg NH_3, 0.2 mg/kg N_2H_4	[14]
pH 7 with Oxygen (Oxygenated Treatment)	6.5 - 7.3	50-200 μg O_2 /kg, conductivity <0.1 μS/cm	[15]
Combined Treatment	8.0-8.5	$NH_3 + O_2$	[16]

The most common feedwater chemistry used in fossil-fired SCW power plants is now OT, which was developed in the 1970's in the former West Germany and subsequently adopted in Russia. In the 1990s, it started to be adopted in the United States as well. A detailed exposition of OT and guidelines for its use can be found in Bursik et al. [17]. The presence of oxygen reduces the corrosion rate of the carbon steel feedtrain piping, reducing corrosion product transport to the boilers. This reduction in corrosion rate has been attributed to a change in the nature of the oxide formed (from magnetite to either hematite or an iron oxyhydroxide). Plants containing copper alloys in the feedtrain cannot use OT because the normally-protective oxide film formed on copper alloys dissolves in high-temperature oxygenated water, resulting in transport and deposition of copper within the core and on the turbines. This water chemistry is compatible with stainless steel components, however, routine inspection and the highest degree of water purity are essential to prevent stress corrosion cracking (SCC).

3.2 High-temperature chemistry monitoring strategies and techniques

In conventional CANDU nuclear power plant (NPP) designs, monitoring of chemistry and corrosion product transport is performed through use of on-line (in-situ and local) measurements and grab samples at selected points throughout the primary and secondary circuits. The direct-cycle design of the SCWR is in a sense a hybrid of these two circuits, similar in concept to a BWR, and will require monitoring at many of the same locations: purification inlets and outlets, feedwater, main "steam", drains, pump outlets and the condenser hotwell. While the optimal water chemistry is yet to be determined, the monitored parameters are likely to be the same as those monitored in existing nuclear plants: pH; conductivity and cation conductivity; concentrations of O_2, H_2, and any additives; concentrations of impurities such as Na, Si and Cl; and concentrations of corrosion and activation products. Not all of these parameters would necessarily be control parameters, but would still be required for identifying and diagnosing problems. The techniques used for measuring many of these parameters will have to be modified and in many cases new techniques will have to be developed to provide meaningful data at high and supercritical temperatures.

Recent advances in monitoring techniques include development of advanced methods for obtaining grab samples and development of high-temperature devices for pH, dissolved H_2 and corrosion monitoring. Grab sample techniques have been criticized for non-representative sampling and deposition along sampling lines [18]; in particular, O_2 can react rapidly with metal surfaces at high temperatures. When sampling particles, the use of isokinetic sampling, which involves the use of advanced sampling nozzles [19] and proper design of sampling lines to ensure turbulent flow [18], can provide more representative samples. However, it has long been recognized that measuring representative corrosion product concentrations in a high-temperature system is a difficult task [20], as the species of interest can interact with the oxide layers on the walls of the sampling line, and their solubilities change as the sample is cooled along the sample line [21].

Chemistry monitors suitable for use at high temperatures have been in development since the early 1990s and include electrochemical noise probes, high-temperature electrodes, dissolved H_2 sensors and microelectrodes. It has been shown that electrochemical noise can be correlated with corrosion rate [22] in high-temperature water, and it may be possible to use this technique to monitor feedtrain and inlet feeder corrosion. High-temperature yttrium-stabilized zirconia (YSZ) membrane electrodes can serve as reference electrodes for electrochemical corrosion potential (ECP) monitoring or as on-line pH electrodes when coupled with Fe/FeO or Ni/NiO [23, 23]. Dissolved hydrogen concentration can also be monitored at the inlet feeders through use of Pd/Pt resistance sensors [25]. Platinum disc microelectrodes could be used to measure pH, dissolved O_2 and H_2 concentrations, as well as the concentrations of metals and some additives (e.g., N_2H_4) [26]. All of these techniques are limited, however, to the low-supercritical region (~400ºC) where electrochemical processes still occur. Monitoring of the chemistry conditions in the gas-like supercritical fluid exiting the core (~620ºC) will be vitally important to SCWR operation, as oxidizing conditions could result from water radiolysis in-core, and techniques for monitoring in this region will have to be developed. Aside from grab samples for corrosion product transport measurements, where existing technology may only need minor modifications, measurements of redox species (H_2 and O_2) will be important in this region, and new and innovative techniques will be needed to minimize chemical reactions along the sampling lines.

Chemistry control techniques will also need revision, and perhaps novel approaches will be needed. Unlike the indirect-cycle design of conventional CANDU reactors, whose steam generators have the serendipitous effects of providing full-flow filtration of primary side corrosion products, physical separation from impurities resulting from condenser ingress, and a phase boundary to limit carryover in the steam, the direct-cycle CANDU-SCWR must deal with corrosion product transport, impurities and carryover by different means (e.g., corrosion resistant materials, condensate polishing and high-temperature purification).

3.3 Areas of concern for chemistry monitoring and control in a SCWR

For illustration, a conceptual layout for a SCWR is presented in Figure 4. Four points of particular concern for chemistry monitoring and control in this SCWR cycle are indicated: (1) condenser/post-condenser, (2) post-deaerator, (3) final feedwater, and (4) post-core/pre-turbines. The condenser (1) can provide a route for ingress of impurities from the cooling water due to tube leaks and air ingress. Sources of particulates in the condensate include drains returns and steam carryover. Fossil-fuel power industry experience suggests that full-flow condensate polishing will be necessary to reduce the concentrations of impurities and corrosion products, especially if, as noted earlier, the oxygenated feedwater treatment is adopted.

The feedwater entering the high pressure (HP) feedwater heaters after leaving the deaerator (2) will also be a location where chemistry control will be important. Just upstream of the core feed pump is an obvious point for chemical dosing, but because the deaerator is often a sink for the drains from the HP feedwater heaters, it is also likely to contain significant quantities of corrosion products. Figure 4 is drawn similar to most BWRs [27], in that the HP drains are cascaded to the LP feedwater heaters, which then cascade to the condenser, providing a route whereby the condensate polisher should remove the corrosion products. Whether additional purification will be necessary at a point past the deaerator is not certain, and will depend on the predicted amount and location of in-core deposition and the maximum allowable deposition on fuel; purification at point (3) may even be necessary. Recently, Kozinski et al. [28] described the available technologies for high temperature-pressure (HTP) purification and found that a combination of several techniques may be used, including the use of hydrocyclones and cross-flow microfiltration.

The chemistry at point (3) is of great importance to minimize corrosion product deposition in-core. The concentrations of corrosion products must be measured at this location, and control of water radiolysis may require addition of hydrogen or some other additive. Kysela et al. [29] have suggested the possible use of a ‘dual’ water chemistry, in which the feedwater and core chemistries are different. An oxygen feedwater chemistry can be converted to a hydrogen chemistry within the core by adding hydrogen upstream of the core, allowing the radiation field to recombine the hydrogen and oxygen stoichiometrically and produce a residual hydrogen concentration to minimize the net radiolytic decomposition of water.

Finally, point (4), the post-core region will require chemistry monitoring, and possibly some chemistry control. If radiation chemistry studies show that the net production of O_2 and H_2O_2 by water radiolysis cannot be controlled through addition of hydrogen or some other additive, the resulting oxidizing conditions may have to be controlled post-core. In the absence of a predictive model of in-core water chemistry, it is not clear at this time how chemistry control at point (4) might be achieved.

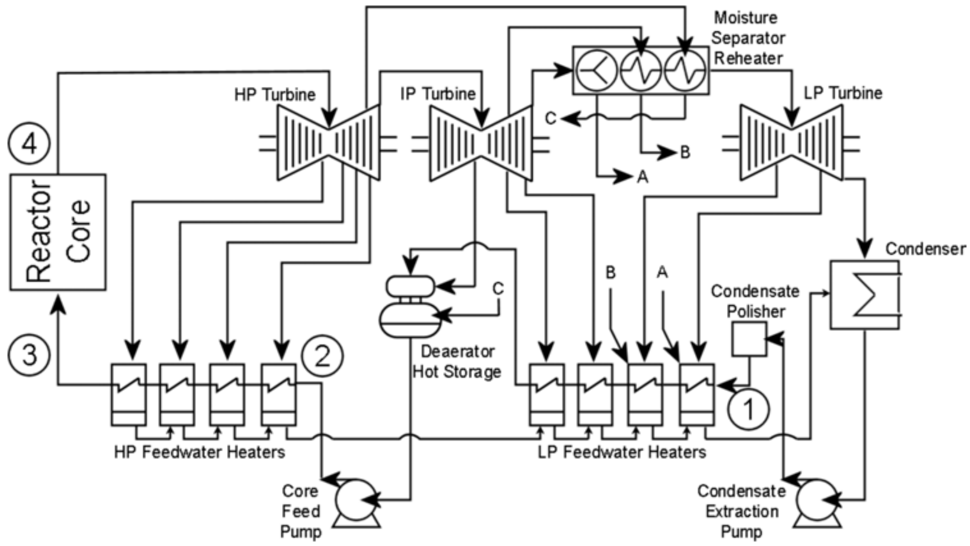

Figure 4 A conceptual layout of a direct-cycle SCWR with steam reheat.

4. Conclusions

The CANDU-SCWR poses many new challenges for chemistry monitoring and control, including especially the challenges of monitoring and controlling chemistry in high-temperature and high-pressure streams. A number of techniques are available for sampling and probing of the relatively low-temperature feedwater circuit, but more research is needed to develop techniques for accurately measuring relevant parameters in the gas-like supercritical fluid exiting the SCWR core. While there are still insufficient data to specify a complete water chemistry regime for a CANDU-SCWR, the evidence suggests that an oxygenated feedwater treatment would be the best feedwater chemistry. A major challenge will be to find an effective means of suppressing the net radiolytic production of oxidizing species in the reactor core. If a suitable means cannot be found, materials degradation could be very high at the core outlet due to the combination of highly oxidizing conditions and high temperatures. This may require chemical addition at the core outlet to reduce the concentrations of oxidizing species.

It is important that chemistry control and system design work together. Alloy selection for all plant components must address both material integrity concerns and corrosion product transport. Consideration should also be given to overall plant layout and system design, as fuel fouling concerns should not come second to gains in efficiency.

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