Kirk-Othmer Encyclopedia of Chemical Technology.

NUCLEAR REACTORS, WATER CHEMISTRY OF LIGHTWATER REACTORS

As of 1994 there were 105 operating commercial nuclear power stations in the United States (1) (see Power generation). All of these facilities were light, ie, hydrogen–water reactors. Seventy-one were pressurized water reactors (PWRs); the remainder were boiling water reactors (BWRs).

In a PWR, a closed circuit of high pressure, high temperature water transfers heat from the reactor core to once-through or recirculating U-tube steam generators, as shown in Figure 1. The steam (qv), which is produced on the secondary side of the steam generator, is used to drive a turbine generator. In contrast to fossil-fired steam-generating equipment, steaming in a PWR occurs on the outside of the boiler tubes, ie, the secondary side, where a large number of tube-to-tube support plate and tube-to-tubesheet crevices exist. Extensive corrosion has been observed in such crevices and beneath sludge piles on the tube–tube support plates and tubesheet (2). Control of secondary chemistry is critical if high concentration of aggressive chemicals and accelerated local corrosion are to be avoided (see Corrosion and corrosion control; Water, industrial water treatment).

Boron, in the form of boric acid, is used in the PWR primary system water to compensate for fuel consumption and to control reactor power (3). The concentration is varied over the fuel cycle. Small amounts of the isotope lithium-7 are added in the form of lithium hydroxide to increase pH and to reduce corrosion rates of primary system materials (4). Primary-side corrosion problems are much less than those encountered on the secondary side of the steam generators.

In a BWR (Fig. 2) steam is generated in the reactor core and used directly to drive a turbine generator. Steam generators are not employed in modern BWRs, although several early units were furnished with such equipment as a source of low pressure steam. Radiation levels during operation near the turbine, condenser, and feedwater heaters are higher than in a PWR as a result of steam transport of short-lived activation products. Other than this consideration, the power-generating cycles of PWRs and BWRs are reasonably similar. No reactivity or pH control additives are used in BWRs. As a result, the corrosion behavior of the materials of construction is dependent primarily on coolant oxygen concentrations which are governed by the radiolytic decomposition rates of water and steam–water equilibrium relations. In some cases, these parameters are controlled by the addition of dissolved hydrogen to the feedwater entering the reactor vessel.

1. Pressurized Water Reactors

1.1. Primary System

In a PWR, reactor coolant is circulated continuously at ca 300°C and 15 MPa (150 bar). Austenitic stainless steel is employed for the main system piping, or cladding, reactor vessel cladding, pressurizer cladding, pumps (qv), valves, and auxiliary piping. The steam generator tubing used in modern U.S. nuclear facilities is Alloy 600, ca 75 wt % Ni, 8 wt % Fe, and 16 wt % Cr. However, most of the tubing in replacement units is Alloy 690, ca 55 wt % Ni, 8 wt % Fe, and 30 wt % Cr. The steam generator tubing accounts for ca 75% of the total area exposed to the primary coolant and Zircaloy 4 and stainless steel account for ca 20 and 5%, respectively (5).

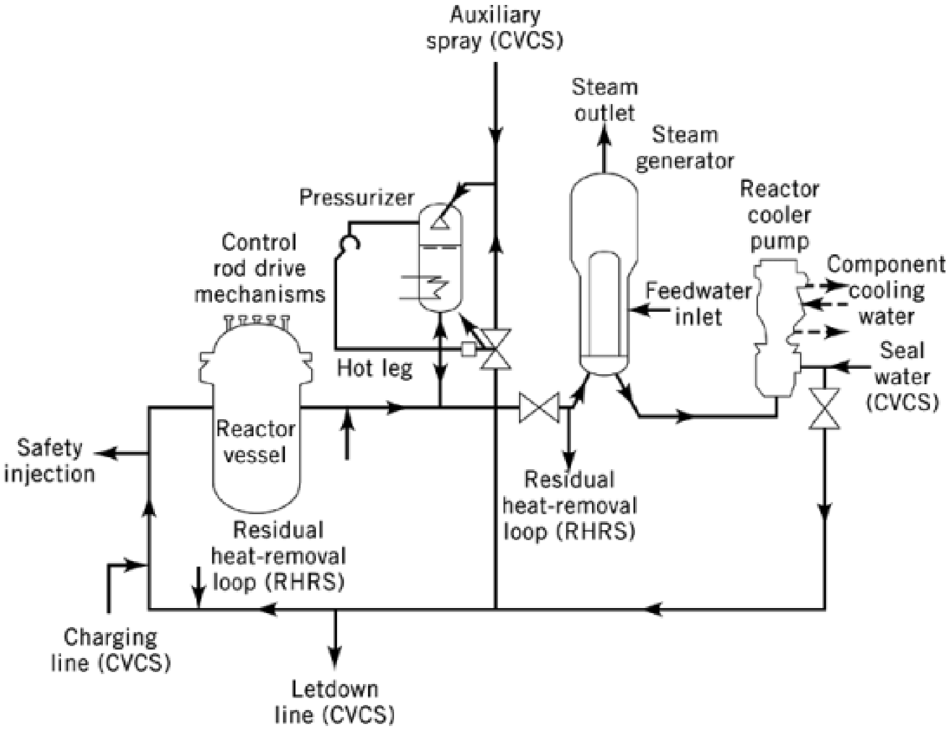

Fig. 1. Pressurized water reactor (PWR) coolant system having U-tube steam generators typical of the 3–4 loops in nuclear power plants. PWR plants having once-through steam generators contain two reactor coolant pump-steam generator loops. CVCS = chemical and volume-control system.

The primary water specifications for a PWR are given in Table 1 (4). Rigid controls are applied to the primary water makeup to minimize contaminant ingress into the system. In addition, a bypass stream of reactor coolant is processed continuously through a purification system to maintain primary coolant chemistry specifications. This system provides for removal of impurities plus fission and activated products from the primary coolant by a combination of filtration (qv) and ion exchange (qv). The bypass stream also is used both to reduce the primary coolant boron as fuel consumption progresses, and to control the ^{7}Li concentrations.

Oxygen is a prime factor in the corrosion of system materials and the release, activation, and redeposition of activated corrosion products. Dissolved hydrogen is maintained to promote rapid recombination of the oxygen whether radiolytically formed or introduced into the coolant from other sources, thereby minimizing corrosion rates.

The quantity of boric acid maintained in the reactor coolant is usually plant specific. In general, it ranges from ca 2000 ppm boron or less at the start of a fuel cycle to ca 0 ppm boron at the end. Most plants initially used 12-month fuel cycles, but have been extended to 18- and 24-month fuel cycles, exposing the materials of construction of the fuel elements to longer operating times. Consequently concern over corrosion problems has increased.

The reactor coolant pH is controlled using lithium-7 hydroxide [72255-97-1], ^{7}LiOH. Reactor coolant pH at 300°C, as a function of boric acid and lithium hydroxide concentrations, is shown in Figure 3 (4). A pure boric acid solution is only slightly more acidic than pure water, 5.6 at 300°C, because of the relatively low ionization of boric acid at operating primary temperatures (see Boron compounds). Thus the presence of lithium hydroxide,

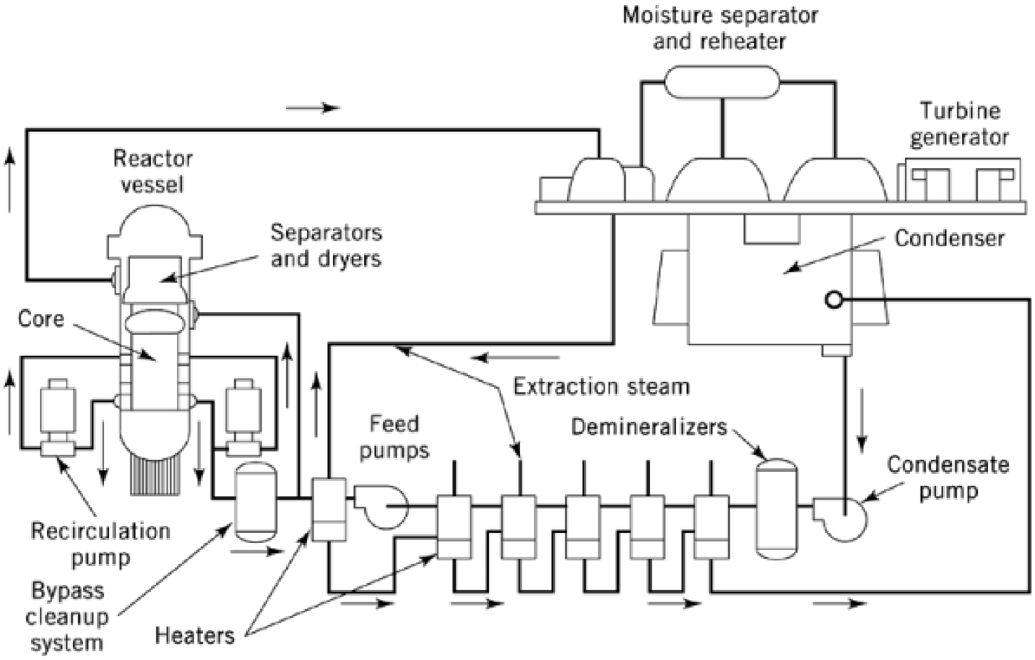

Fig. 2. Boiling water reactor (BWR) system.

Table 1. PWR Primary System Specifications[a, b]

Parameter[c]	Normal value
O_2, ppb	<5
H_2,[d] mL/kg H_2O	25–50
Cl, ppb	<50
F, ppb	<50
SO_4, ppb	<50
suspended solids, ppb	<350
SiO_2, ppb	[e]

[a] Ref. 4.
[b] Values are for normal power operation. Conductivity, pH, and concentrations of lithium and boron are plant specific and vary over the fuel cycle according to the control scheme used. See Fig. 3.
[c] ppb = parts per billion.
[d] At STP.
[e] Monitored in fresh makeup water. Should be $<$ 100 ppb.

which has a much higher ionization, increases the pH ca 1–2 units above that of pure water at operating temperatures. This leads to a reduction in corrosion rates of system materials (see Hydrogen-ion activity).

The pH control scheme is plant specific. It is selected on the basis of impacts on fuel and materials integrity and radiation field control. The pH at 300°C should be maintained above 6.9, and from a radiation control point of view, at 300°C a pH up to 7.4 is desirable. However, the high lithium values, required for pH values between 6.9 and 7.4 at 300°C, during the initial part of the fuel cycle, have the potential of accelerating the corrosion of Zircaloy fuel cladding and of increasing the susceptibility of mill-annealed Alloy 600 tubing used in some recirculating steam generators (RSGs) to primary-side stress corrosion cracking (4, 6, 7).

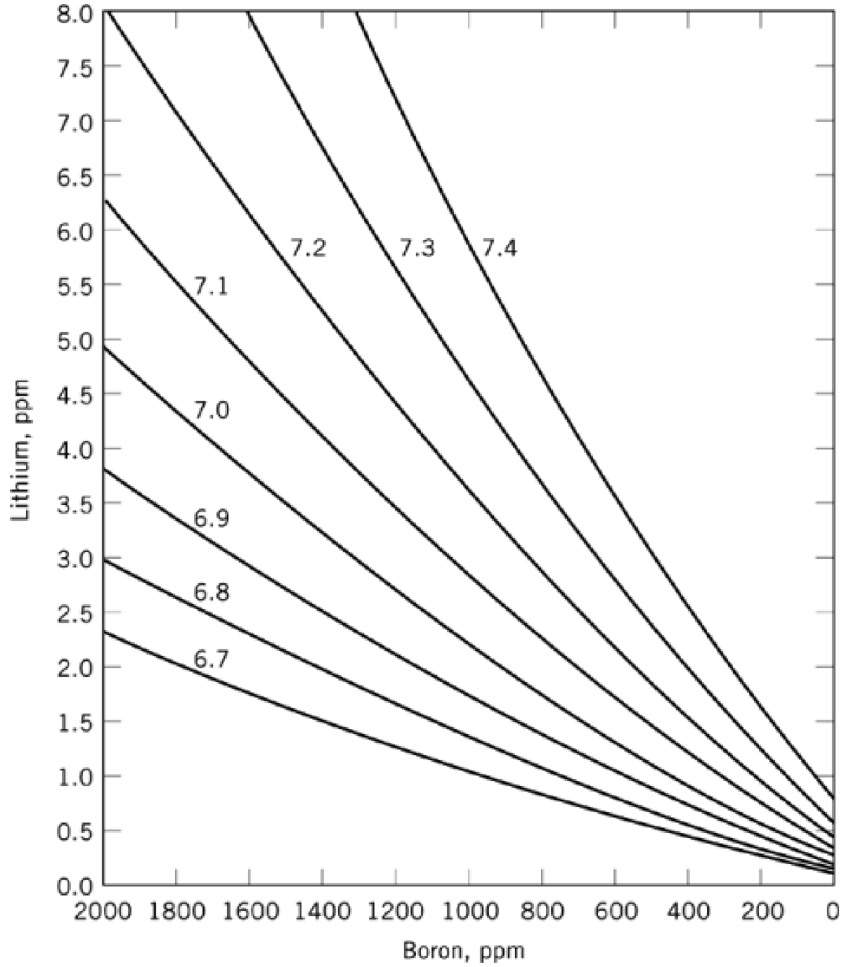

Fig. 3. Lithium hydroxide–boric acid relationships for various pH values at 300°C (4).

In some plants high silica, SiO_2, levels (up to the ppm range) have been found in the primary water. The source is the boroflex material used in the fuel storage racks in the spent fuel pool. The SiO_2 leaches into the pool water, then finds its way into the primary system when the primary water comes in contact with the pool water during refueling and similar maintenance. The SiO_2 can only be effectively removed by reverse osmosis (qv).

A schematic of the radiation contamination for a PWR is shown in Figure 4. Whereas numerous attempts have been made to model this process, there has been only limited success (8–11). Trends in the radiation field regarding time for RSG channel heads are shown in Figure 5 (12). The fields vary over a wide range, but reach a peak between four and six years. The two principal nuclides leading to out-of-core shutdown radiation levels are cobalt-58 [13981-38-9], ^{58}Co, and cobalt-60 [10198-40-0], ^{60}Co, (4, 5) (Table 2). Studies show that electropolishing of the plenum surfaces of RSGs before they go into service can reduce the deposition rate of cobalt, and consequently, the rate of radiation buildup (13). The electropolishing smoothes the microscopic surfaces of the plenums and reduces the sites where the cobalt can deposit.

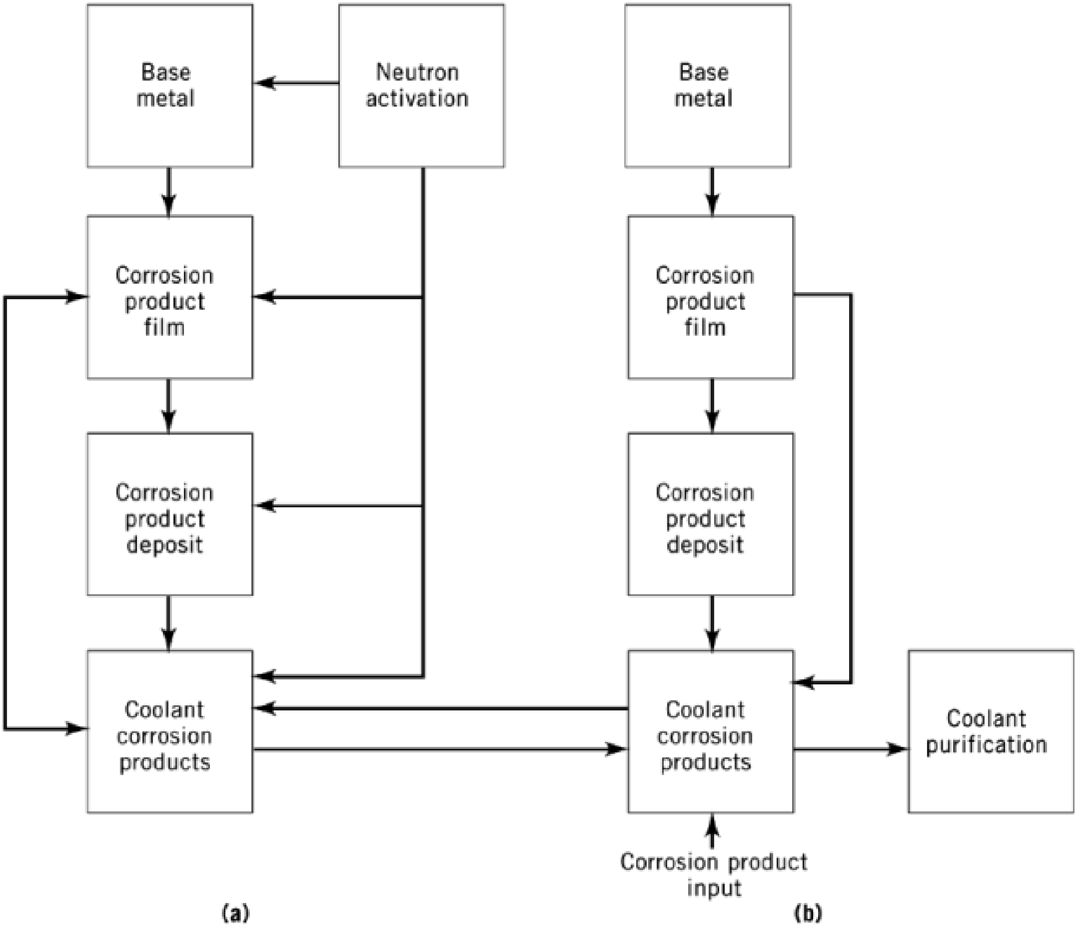

Fig. 4. Contamination process schematic for PWR where (**a**) is inside the core and (**b**), outside the core.

Table 2. Corrosion Products Contributing to PWR Shutdown Radiation Levels[a]

Isotope	Half-life	Source	Gamma rays		Disintegration, MeV
			Energy, MeV	Abundance, %	
^{58}Co	71 d	$58_{\mathrm{Ni}(n,p)}$	0.511	0.3	0.98
			0.810	0.99	
			0.865	0.01	
			1.67	0.01	
^{60}Co	5.27 yr	$59_{\mathrm{Co}(n,\gamma)}$	1.17	1.0	2.5
			1.33	1.0	

[a] Ref. 3.

1.2. Secondary System

The water quality specifications for the feedwater and blowdown water in a recirculating steam generator (RSG) and the feedwater for a once-through steam generator (OTSG) are given in Table 3 (14).

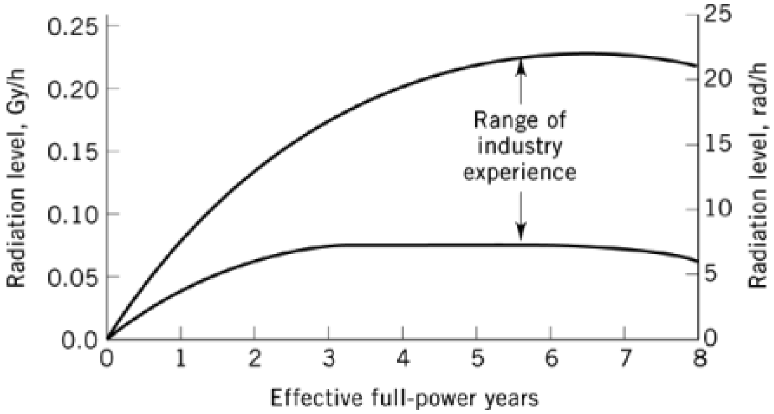

Fig. 5. RSG channel head shutdown radiation fields (12).

1.2.1. Recirculating Steam Generator

The corrosion performance of many RSGs in commercial power stations in the United States has been marginal (2). Many tube bundles have had to be replaced. Many tubes have been plugged or sleeved with inserts as a result of excessive corrosion on the secondary side.

Sodium phosphate treatment was originally used for pH control and conditioning of boiler water (15). Initially, a boiler water concentration range, but not a sodium-to-phosphate ($Na:PO_4$) molar ratio, was specified. Some utilities opted to control the phosphates at various levels within the allowable range (16, 17). During condenser in-leakage, the phosphate and calcium ion reactions depleted the steam generator water of soluble phosphate, increasing the $Na:PO_4$ molar ratio and forming free caustic. This resulted in an increased rate of Alloy 600 steam generator tubing stress corrosion cracking. Although recommended phosphate control parameters were modified, corrosion attack continued. For example, wastage or local wall thinning was observed. An all volatile treatment (AVT) employing ammonia (qv) was then used for pH control of the feedwater. As of this writing (ca 1995) the treatment of choice is AVT (Table 3).

Shortly after the conversion to AVT, inward deformation of the Alloy 600 tubing at the tube-to-tube support plate interface region was detected. This phenomenon, commonly referred to as denting (18), results from excessive corrosion of carbon steel in the crevice region between the tube and the tube support plate; as denting progresses, tube support plate cracking can occur. The tube stress corrosion cracking also continued. Newer support plate materials and modified support plate designs were used in later units and in replacement units. In denting, the accelerated corrosion of the carbon steel tube support appears to result from the ingress of chlorides and other contaminants, such as O_2 and Cu^{2+}.

Stress corrosion cracking, prevalent where boiling occurs, concentrates corrosion products and impurity chemicals, namely in the deep tubesheet crevices on the hot side of the steam generator and under deposits above the tubesheet. The cracking growth rates increase rapidly at both high and low pH. Either of these environments can exist depending on the type of chemical species present.

Possible remedial and preventive actions are as follows (14). (*1*) Use of elevated hydrazine [302-01-2], N_2H_4, treatment where hydrazine in the range of several hundred ppb is added to the feedwater thus producing a reducing atmosphere in the steam generator. (*2*) Boric acid treatment where boric acid in the range of 5–10 ppm B is added to the feedwater. This reduces the growth rate of corrosion products that cause denting and possibly helps arrest tube stress corrosion cracking. (*3*) Alternative amine treatment where, for example, morpholine [110-91-8] or ethanolamine [141-43-5] (ETA), is added in place of ammonia to the feedwater to produce a more uniformly basic environment throughout the secondary system. The transport of corrosion

Table 3. PWR Steam Generator Water Specifications[a, b]

Parameter[c]	Once-through feedwater	Recirculating	
		Feedwater[d]	Blowdown water
components, ppb			
N_2H_4	20[e]	<100	
O_2	≤ 3	<5	
Na	≤ 3		ca 2
Cl	≤ 5		ca 2
SiO_2	≤ 10		
total Fe	≤ 5	<5	
total Cu	≤ 1	<1	
SO_4	≤ 3		ca 3
other	[f]	[g]	[g, h]
cation conductivity at 25°C, μS/cm			
corrected[i]	≤ 0.2	≤ 0.2	
ammonia AVT[j]			0.15
nonammonia AVT[j]			0.5

[a] Ref. 14.

[b] Values correspond to normal power operation.

[c] ppb = parts per billion.

[d] The pH is plant specific, depending on additive used and secondary system materials. Feedwater generally should be equivalent to pH = 9.3 at 25°C for ammonia and carbon steel equipment.

[e] Value given is minimum.

[f] Organics related to use of amines for pH control. Value is plant specific.

[g] Boron related to boric acid treatment of $\sim 5 - 10$ ppm B in feedwater.

[h] For caustic crevice environment, a plant-specific chemical impurity molar ratio <0.5 is defined, eg, Na:Cl molar ratio <0.5.

[i] Corrected for the presence of organics such as acetates and formates.

[j] AVT = all-volatile treatment.

products into the steam generators is thus reduced. (*4*) Molar ratio management where the ratio of cations to anions in the steam generator water is controlled reducing the possibility of forming environments in boiling crevices and underdeposit areas which are conducive to accelerating the stress corrosion cracking (SCC) of Alloy 600. For caustic SCC environments the desired molar ratio, eg, Na:Cl, is <0.5.

Before any remedial or preventive actions are implemented, an evaluation should be conducted as to applicability to the specific plant. The evaluation should continue while the actions are in progress. The main action should be to take measures to reduce the ingress of contaminants into the steam generator by using more reliable materials, such as in the condenser tubes, to reduce leakage. Contaminant control equipment, such as full-flow condensate demineralizers, should also be employed.

1.2.2. Once-Through Steam Generator

The corrosion of OTSGs has not been as extensive or as serious a problem as that of RSGs. Design and operating differences between these systems may be responsible for the corrosion differences. For example, the main fraction of impurities entering the OTSG via the feedwater is transported from the OTSG by the steam. In RSG designs, impurities concentrate in the bulk water. Moreover, full-flow condensate demineralizers, which significantly reduce impurity ingress into the system, especially from condenser tube leaks, are employed at all OTSG PWRs. Condensate demineralizers are not employed at most RSG PWRs.

Feedwater quality control is the main method for controlling the chemistry environment in OTSGs (Table 3). Whereas the all-volatile treatment (AVT) utilizing ammonia was used first, a switch has been made to morpholine or other amines for better distribution in the steam plant (14). The amount of corrosion products (mainly iron) carried into the OTSGs has been greatly reduced.

Most of the Alloy 600 outer diameter tube corrosion has occurred in the region of the upper tubesheet near the open lane, ie, an untubed lane across the middle of the steam generator (16, 17). The steam carries entrained droplets of water through the open lane to the upper tubesheet region where the droplets dry out and concentrate the chemicals. Long tube inserts have been used to sleeve tubes in this region where wall defects have been detected.

OTSGs also experience deposition of material on the flow areas in the tube support plates which causes an increase in pressure drop and eventual reductions in plant power production.

The possible remedial and preventive actions are hot soaks and drains during cooldown to help remove soluble deposited material, chemical cleaning to remove corrosion products and reduce the pressure drop (see Metal surface treatments), and reduced corrosion product transport into OTSG using amines other than ammonia in feedwater (14).

2. Boiling Water Reactors

BWRs operate at ca 7 MPa (70 bar) and 288°C. Some of the coolant passing through the core is converted into steam which is separated from the water with equipment inside the reactor vessel (see Fig. 2). The steam goes to the turbine generator while the water is recirculated back to the bottom of the core. A side stream is continuously purified using demineralizers and filters to control the water quality of the reactor water. Full-flow condensate demineralizers are also used to control the ingress of impurities into the reactor water from the steam plant.

The BWR water chemistry parameters are given in Table 4 (19). Originally, no additives were made to feedwater–condensate or the primary water. The radiolytic decomposition of the fluid produced varying concentrations of O_2 in the reactor vessel, ranging from about 200 ppb O_2 in the reactor recirculation water to about 20 ppm O_2 in the steam. Stoichiometric amounts of hydrogen were also produced, ie, 2 mL H_2 for each mL of O_2. Feedwater O_2 was about 30 ppb, hence the radiolytic decomposition of the water was a primary factor in determining the behavior of materials in the primary system and feedwater systems.

Some of the earlier BWR units had feedwater heaters having copper alloy tubes. The environment of high oxygen and neutral pH water led to high copper concentrations in the feedwater and to undesirable deposits on the fuel and inlet fuel nozzles (20). In some instances, the copper deposits resulted in an increase in core pressure drop and necessitated plant power reduction. The copper alloys were eliminated from the feedwater system in subsequent plants and most existing plants.

Many instances of intergranular stress corrosion cracking (IGSCC) of stainless steel and nickel-based alloys have occurred in the reactor water systems of BWRs. IGSCC, first observed in the recirculation piping systems (21) and later in reactor vessel internal components, has been observed primarily in the weld heat-affected zone of Type 304 stainless steel.

Three factors are important in such cracking: total stresses, sensitization of heat-affected weld zones, and chemical species such as oxygen and hydrogen peroxide in the water. In some cases for recirculation piping, induction heating of welds subsequent to fabrication has been used to change the stress of the inner pipe wall from tensile to compressive. This change reduces the likelihood of IGSCC. Type 316NG stainless steel has been used in replacement and repair situations to eliminate the tendency for sensitization.

Laboratory experiments have shown that IGSCC can be mitigated if the electrochemical potential (ECP) could be decreased to -0.230 V on the standard hydrogen electrode (SHE) scale in water with a conductivity of 0.3 μS/cm (22). This has also been demonstrated in operating plants. Equipment has been developed to monitor

Table 4. BWR Normal Water Chemistry Values[a, b]

Parameter[c]	Median value
Reactor water	
conductivity at 25°C,μS/cm	ca 0.11
Cl, ppb	ca 1
SO_4, ppb	ca 2
Zn, ppb	5–10[d]
electrochemical potential, V	[e]
O_2 in recirculation water, ppb	[f]
SiO_2, ppb	$<$ 100
Feedwater / condensate	
feedwater conductivity at 25°C, μS/cm	0.06
feedwater total Fe, ppb	ca 1–3[g]
feedwater total Cu, ppb	ca 0.1–0.2[g]
O_2, ppb	ca 30

[a] Ref. 19.

[b] Values given correspond to normal power operation.

[c] ppb = parts per billion.

[d] Consistent with plant program for zinc injection.

[e] Corrective action should be initiated when value is > -0.23 V against the standard hydrogen electrode (SHE). Plant-specific values should be established for protection of stainless steels and nickel-based critical components.

[f] Plant specific. In range of 200 ppb without H_2 addition and in range of 10 ppb with H_2 addition.

[g] Values depend on plant-specific design and operating procedures for steam plant.

ECP in the recirculation line and in strategic places such as the core top and core bottom, in the reactor vessel during power operation.

The desired ECP value is not obtainable unless H_2 is injected into the feedwater. For example, using no hydrogen addition, the recirculation line O_2 is in the range of 200 ppb and the corresponding ECP is in the vicinity of zero volts. The amount of H_2 required is plant specific. Experiments at one BWR indicated that the required feedwater H_2 concentration ranged from 2.4 ppm in the recirculation line, to 1.4 and 1.9 in the core bottom and top, respectively (23). Measurements at the same relative location for several plants indicate that required H_2 ranges between 0.4 and 1.6 ppm (22). Some plants use material specimens inside the reactor vessel and recirculation line to monitor the effectiveness of the ECP control by measuring the crack growth rate.

Whereas addition of hydrogen to feedwater helps solve the O_2 or ECP problem, other complications develop. An increase in shutdown radiation levels and up to a fivefold increase in operating steam plant radiation levels result from the increased volatility of the short-lived radioactive product nitrogen-16, ^{16}N, (7.1 s half-life) formed from the coolant passing through the core. Without H_2 addition, the ^{16}N in the fluid leaving the reactor core is in the form of nitric acid, HNO_3; with H_2 addition, the ^{16}N forms ammonia, NH_3, which is more volatile than HNO_3, and thus is carried over with the steam going to the turbine.

Figure 6 (24) is a flow diagram of radioactive contamination in BWR. Although iron is the main corrosion product entering the reactor water, as for PWRs, ^{60}Co is the long-term source of shutdown radiation in BWRs. Some operating BWRs were noted to have shutdown radiation levels substantially lower than those of other plants having similar operating conditions and water chemistry. The common denominator for these low radiation levels was the use of tubes containing zinc in the condensers. Confirmation was made via subsequent studies where zinc oxide was added to the feedwater to produce zinc concentrations in the range of 5–10 ppb in the reactor water (Fig. 7) (12). It appears that whereas the Zn inhibits the deposition of cobalt, there are also some side effects. Zinc addition produces ^{65}Zn hot spots within the plant. Zn-65 has a 244 d half-life.

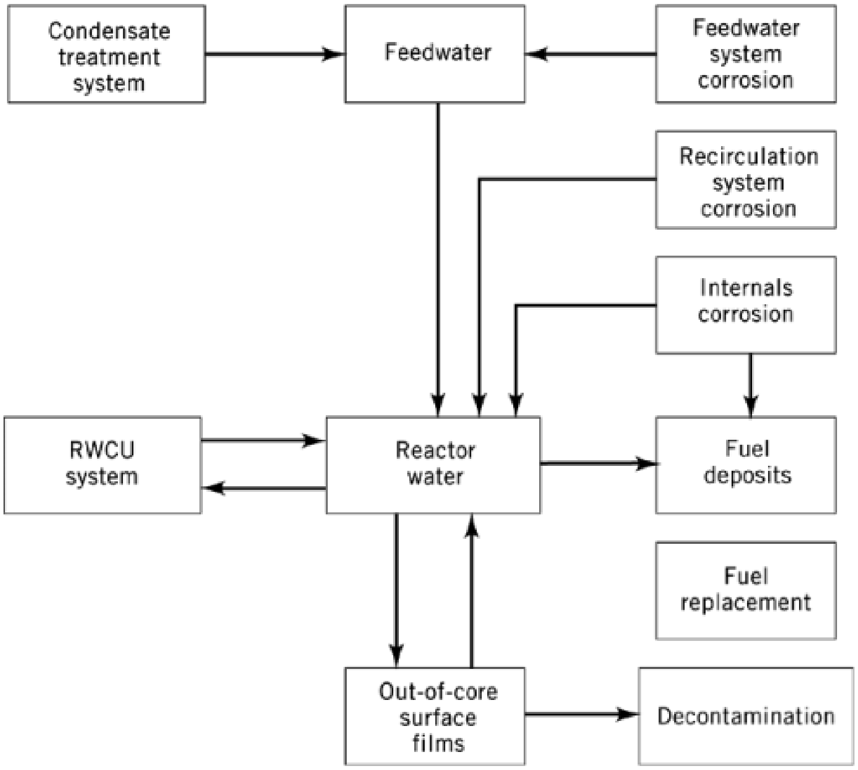

Fig. 6. Impurity flow paths of BWR radioactive contamination (24). RWCU = reactor water cleanup system.

The cobalt deposition rate on new, replacement, or decontaminated recirculation piping surface has been reduced by pretreating the piping using an atmosphere of oxygenated wet steam to form an oxide film (25). Studies have been conducted for both PWRs and BWRs to reduce the cobalt content of materials used in the nuclear parts of the plants, particularly in hardened and wear surfaces where cobalt-base alloys ($\approx 50\%$ Co) are used (26). Some low cobalt materials have been developed; however, the use of the materials is limited to replacement parts or new plants.

3. Economic Aspects

Water chemistry is important to the safe and reliable operation of a nuclear power plant. Improper conditions can lead to equipment and material failures which in turn can lead to lengthy unscheduled shutdown periods for maintenance (qv) and repair operations. Water chemistry can also have an impact on the radiation levels during both power operations and shutdown periods. These affect the ability of personnel to perform plant functions.

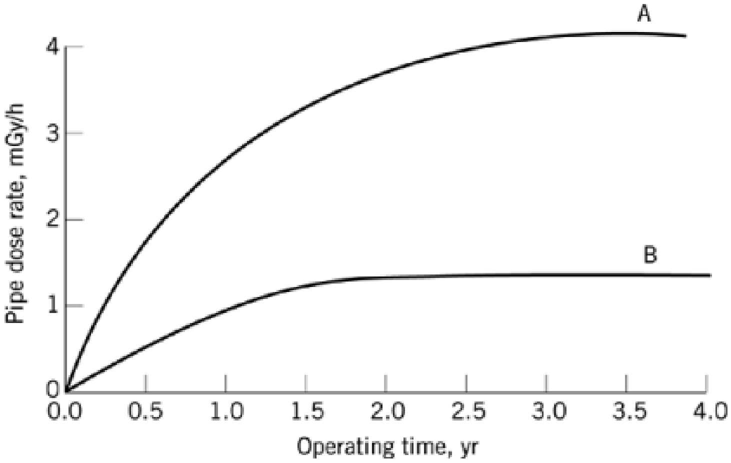

Fig. 7. Radiation buildup at BWRs using normal water chemistry, A, and zinc additions, B (12). To convert mGy to mrad, multiply by 100.

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