Characteristics of Water Chemistry in Heavy Water Recovery System of Nuclear Power Plant

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Abstract: The moisture inside the coolant and moderator system areas of Candu-type nuclear power plant is recycled to prevent the deuterium of value from being lost and the tritium of harm from being spread to the natural environment. The deuterium is separated from the moisture by the successive processes of adsorption and desorption on molecular sieve, cooling with water, purification with activated carbon filter and ion exchange, and evaporation in the deuterium recovery system that consists of dryers, storage tanks, filter, three ion exchange beds, evaporators, and up-grader. The water collected in the dryers is transformed from the kinds of moisture generated inside the primary system of nuclear plant and supplied with the moisture in the fresh air. In addition, hydrazine can be included in the moisture such that the water might be alkaline.

Unexpectedly, the solution in the storage tank followed by dryers is acidic and the deuterium water collected in the up-grader is also acidic in Wolsung nuclear unit 1. The concentration of chloride is some 500 ppm and pH is 3.8 in the storage tanks of dryers, while the white precipitates of aluminum and silica oxides exist to some degree before filter and to fair extent after ion exchange bed. Hydrazine and its derivative, ammonia are not high in the concentration(less than 1ppm). The source of chloride was salt included in the fresh air and the sources of aluminum and silica oxides were products thermally decomposed from molecular sieve. The impurities decrease the removal efficiency of filtration and ion exchange and shorten the operation lifetime of their facilities. In this case the chloride can be transferred to the up-grader in which the packing material, copper, might be corroded. Therefore the ingress of salt in the incoming fresh air should be completely prevented to diminish the breakdown of molecular sieve and to increase the purification capacity of the activated carbon filter and ion exchange bed. The runtime of ion exchange towers was extended more than 30% with the bed arrangement of anion-mixed-mixed ion exchange resins and the cation-to-anion exchange resin ratio of 1:1 on the equivalent basis.

Key-World: Deuterium recovery, Ion exchange, Breakthrough time, Cation, Anion, Resin ratio

1. Introduction

The moisture inside the coolant and moderator system areas of Candu-type nuclear power plant is reused to prevent the deuterium of value from being lost and the tritium of harm from being spread to the natural environment. The deuterium is separated from the moisture by the successive processes of adsorption and desorption on the molecular sieve, cooling with water, purification with activated carbon filter and ion exchange, and evaporation in the deuterium recovery system that consists of dryers, storage tanks, filter, three ion exchange beds, evaporators, and up-grader. The evaporation takes place on the copper packing in the up-grader so that the purity of deuterium recycled may depend on the area of interface between liquid and vapor phases. The up-grader improves the purity of deuterium more than 99.9 wt% according to the difference in the relative volatility of heavy and light water.

The impurities included in the deuterium of low grade may cause the lifetime of he purification system to be short and the purification media to be frequently replaced. The ion exchange resin and the activated carbon filter are supposed to removes the ionic and colloidal substances, respectively, which are partly radioactive. Because they are not reusable, their frequent replacement may result in the increased radioactive waste, the increased treatment expense, the increased radiation exposure to workers, and the increased purchase cost. The objectives of this study were to investigate the source and composition of impurities in the deuterium recovery system and the effect of cation-to-anion resin ratio on the efficiency of ion exchange beds.

2. Deuterium Recovery System

Figure 1 shows the deuterium recovery system for the coolant and the moderator which is made up of the collection, cleanup, and upgrade equipments.

Under the normal condition, it was expected that pH is higher than 7 and chloride is low in concentration. However, the samples contained a lot of white precipitates and the concentration of chloride was so high that pH was low: pH, [Cl], and [Al] are 3~4, 1500ppm, and 700ppm at the reservoir tank behind the Dryer, respectively. Figure 2 shows the data for pH, conductivity, [Cl], [Na], [Al], and [Si]. The white precipitate was generated from the thermal composition of zeolite and NaCl was come from the seawater. The reason why the concentration of Na was lower than that of Cl might be the ion exchange of Na with H in the zeolite.

3. Ion Exchange

The column ion exchange experiment was performed with the simulated water and with the apparatus as shown in Table 1 and Figure 3. Figure 4 and 5 illustrate the pH and conductivity for two different cases of water quality at the outlet of three columns in which mixed, mixed, and anion exchange beds were successively run. Before the breakthrough points, the effluent is neutral and the conductivity is low, while after the breakthrough points, pH and the conductivity are identical to those of the influent. For two different electrolytes, the breakthrough point at the third column was the longest at the cation-to-anion resin ratio of 1:1.

Reference

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Exchanges:The Effect of Electrolyte Anions on Selectivity and Adsorption Capacity, J. of KSEE, 4(3), 195~206, 1999

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Table 1. Electrolyte Composition and Concentration

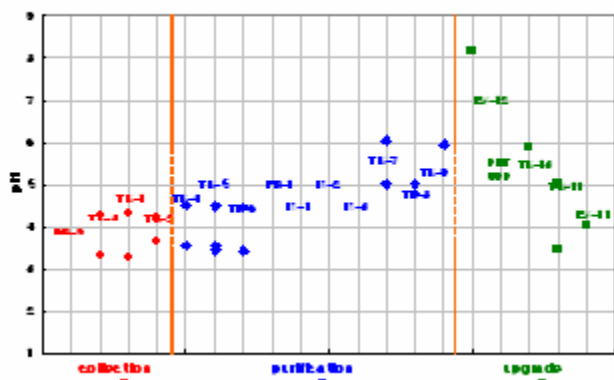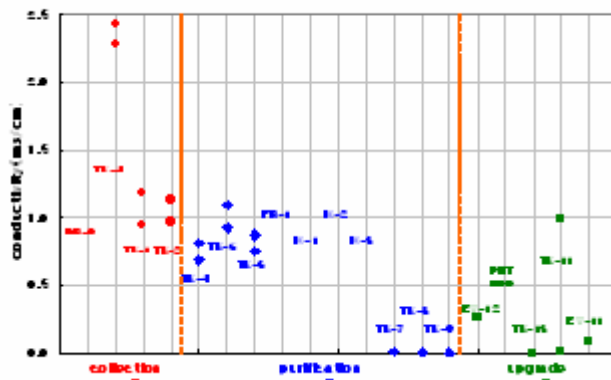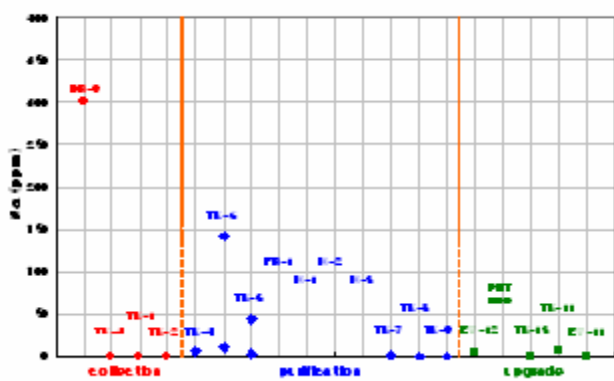|		Cl 315 (ppm)	Cl 315 (ppm)			Cl 315 (ppm)	Cl 315 (ppm)
input(g)	NaCl	4.8024	4.662	concentration(M)	pH	4.54	3.21
input(g)	$CaCl_2$	0.036	0.108	concentration(M)	conductivity(/)	935	924
input(g)	$MgSO_4$	0.036	0	concentration(M)	SS(ppm)	0	0
input(g)	Na_2SiO_3	0.117	0.324	concentration(M))	H^+	2.88×10^{-5}	6.17×10^{-4}
input(g)	$AlCl_3$	2.7	2.556	concentration(M)	Na^+	5.44×10^{-3}	5.44×10^{-3}
input(g)	Na_2CO_3	0.711		concentration(M)	Mg^{2+}	1.66×10^{-5}	0
input(g)	$CuCl_2$	0.3654	0.378	concentration(M)	Ca^{2+}	1.80×10^{-5}	5.41×10^{-5}
input(g)	NaF	0.018	0.54	concentration(M)	Al^{3+}, Cu^{2+}, Ni^{2+}, Fe^{3+}		
input(g)	KCl	0.018	0.09	concentration(M)	OH^-	3.47×10^{-10}	1.62×10^{-11}
input(g)	$FeCl_3$	0.432	0.234	concentration(M)	Cl^-	8.87×10^{-3}	8.87×10^{-3}
input(g))	$NiCl_2$	0.153	0.27	concentration(M)	F^-	2.38×10^{-5}	7.14×10^{-4}
input(g)	$Al(NO_3)_3$	1.8144	1.08	concentration(M)	NO_3^-	1.42×10^{-3}	8.45×10^{-4}
input(g)	Al_2O_3	0/20/60	0	concentration(M)	SO_4^{2-}	1.66×10^{-5}	2.33×10^{-4}
input(g)	SiO_2	0/20/60	0	concentration(M)	CO_3^{2-}	3.73×10^{-4}	0
input(g)	Fe_3O_4	0/5/15	0	concentration(M)	SiO_2		
input(g)	CuO	0/5/15	0	concentration(M)			
input(g)	H_2SO_4	0	0.4119	concentration(M)			
input(g)	HCl	0	0.1855	concentration(M)			
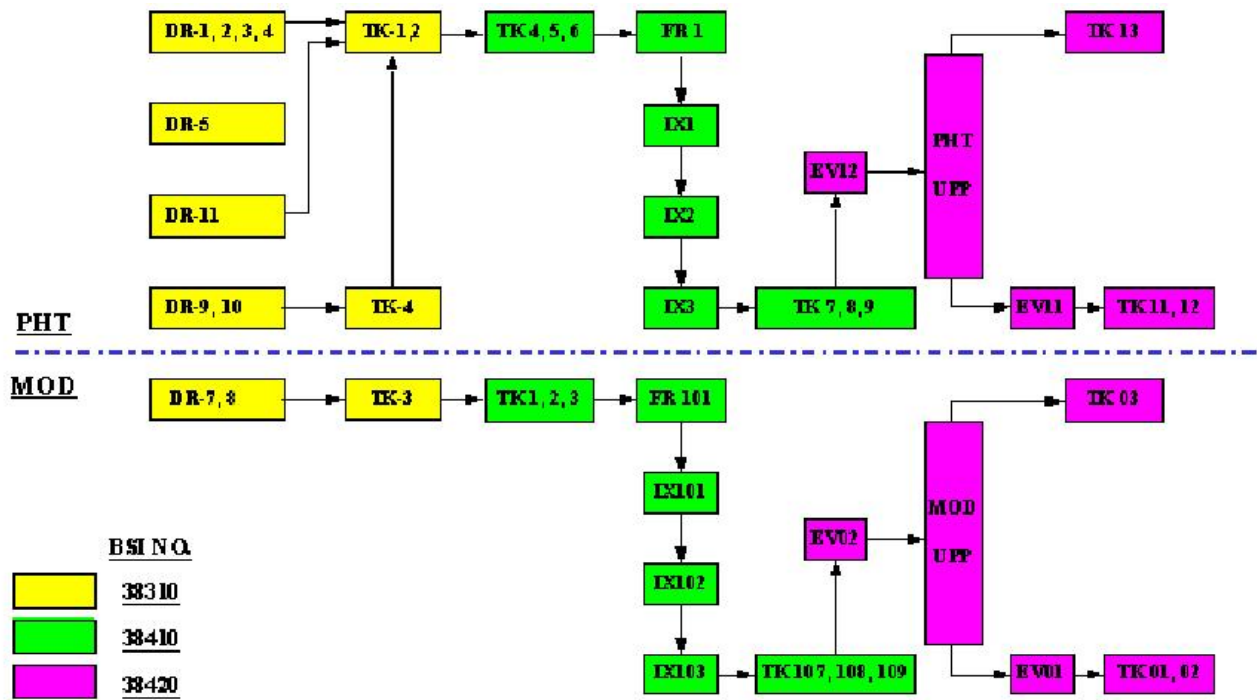
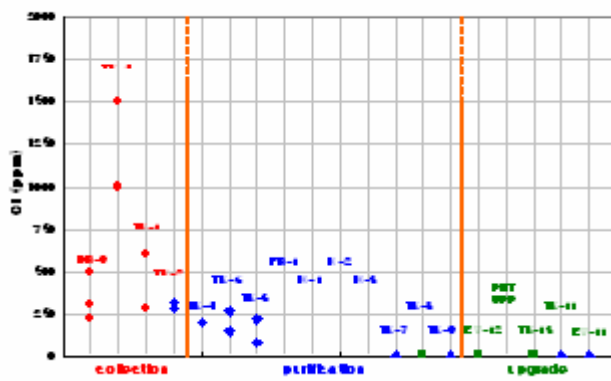Figure 1. Deuterium Purification System : Collection, Cleanup, and Upgrade Facilities
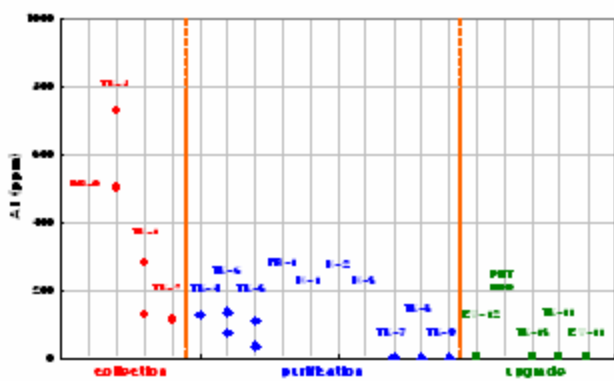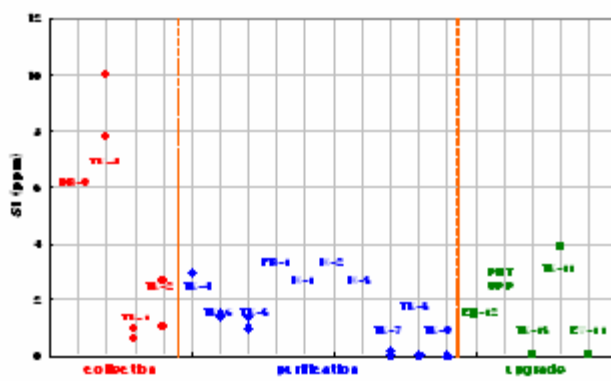
(a) pH

(b) Conductivity

(c) Na

(d) Cl

(e) Al

(f) Si

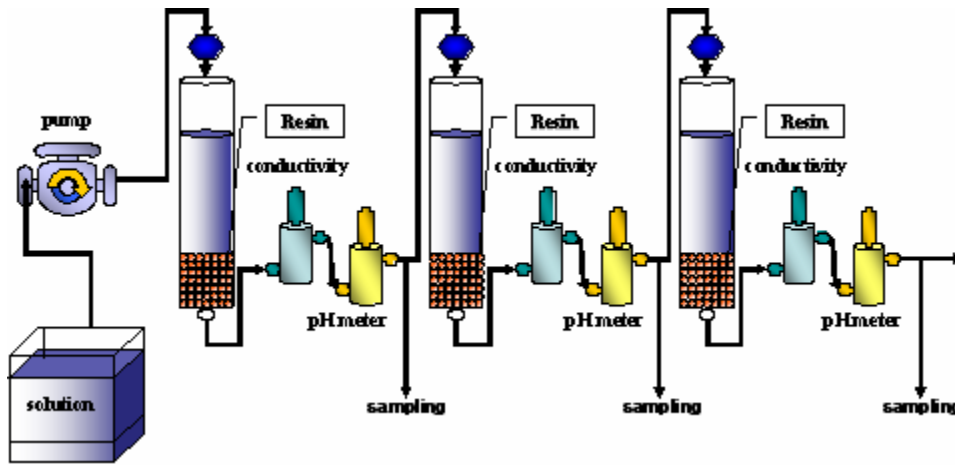Figure 2. pH, Conductivity, Cl, Na, Al, Si in the deuterium recovery system
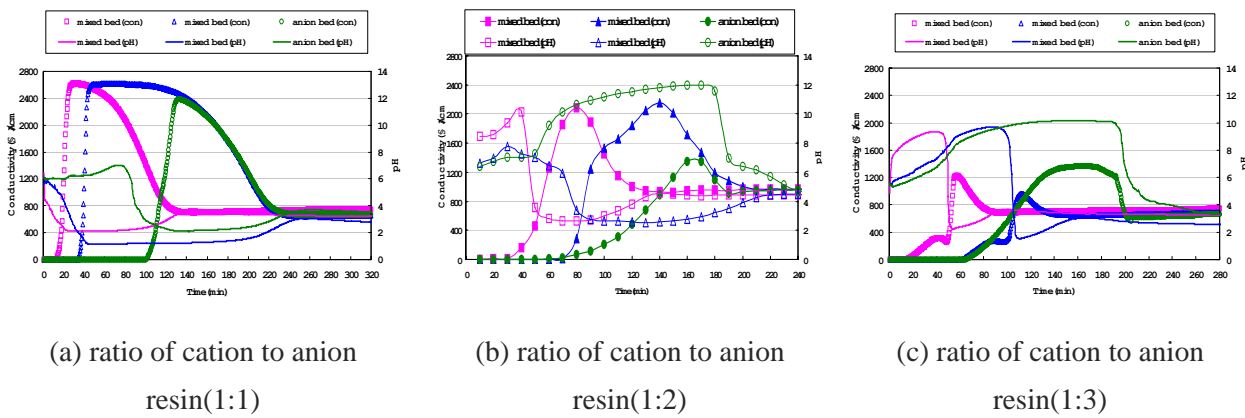
Figure 3. Column Exchange Apparatus

(a) ratio of cation to anion
resin(1:1)

(b) ratio of cation to anion
resin(1:2)

(c) ratio of cation to anion
resin(1:3)

Figure 4. pH and conductivity at the outlet of column with time (Na$^+$=125ppm, Cl$^-$=315ppm, pH=4.5)

(a) ratio of cation to anion resin
(1:1)

(b) ratio of cation to anion resin
(1:2)

(c) ratio of cation to anion resin
(1:3)
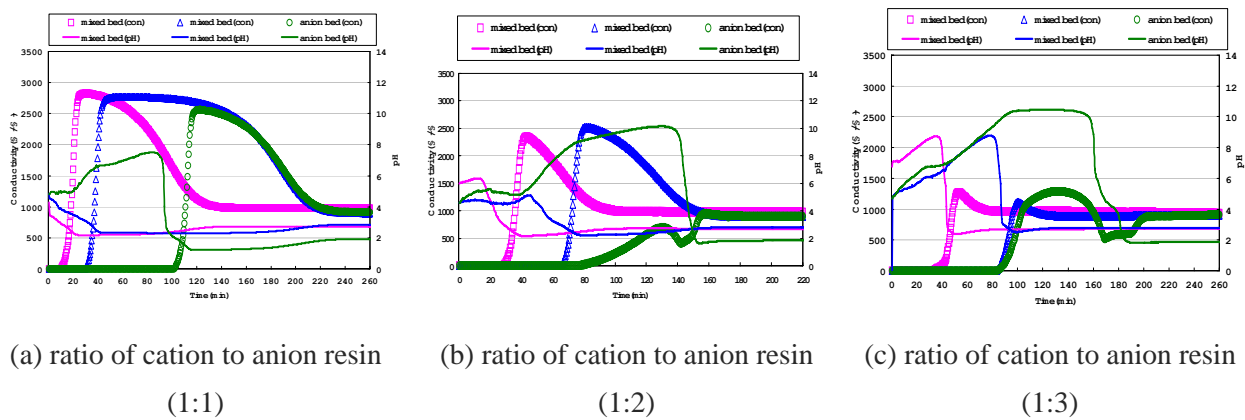
Figure 5. pH and conductivity at the outlet of column with time (Na$^+$=125ppm, Cl$^-$=315ppm, pH=3.2)