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Effect of Different Reduction Methods on the Efficiencies in the Chemical Decontamination Processes

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Abstract−Chemical decontamination is considered to be an effective method for reduction of radiation level by dissolution of radioactive corrosion products and metal oxidizing films existing in the primary system of a nuclear power plant. In this study, the process efficiencies of two chemical decontamination processes (Methods 1 and 2) having different reduction steps were investigated through the operation of a semi-pilot scale decontamination equipment as a continuous work. The reduction step for Method 1 employed an adsorbent with an oxygen source, while a reductant (oxalic acid) was used in the reduction step for Method 2. The dissolution and removal efficiencies of metal species and organic compounds in Method 2 were higher than those in Method 1, implying that oxalic acid in the reduction step increased the process efficiency, their complexes of metal species easily being removed in the decomposition/cleanup step. It was shown that the process employing chemical reduction showed higher dissolution and removal efficiencies rather than the process by the physical adsorption on the adsorbent surface through decontamination processes with different reduction step.

Key words: Adsorption, Chemical Decontamination Process, Oxalic Acid, Reduction

INTRODUCTION

After various periods of normal operation in the primary systems of a nuclear power plant consisting of stainless steels and nickel-based alloys, a coherent oxide layer is usually formed on the surfaces of the components, tubes, and equipment that are in contact with the primary side coolant [Kim et al., 2003a]. The outer part of this oxide layer consists of mainly magnetite (Fe_3O_4) and occasionally some hematite (α-Fe_2O_3). Meanwhile, the inner layer, next to the bulk metal, is a non-stoichiometric mixed oxide having spinel structures. The Fe-chromite ($FeCr_2O_4$) and their other forms substituted by Co and Ni are generated in the inner region and are extremely insoluble even at a high temperature, as long as the reductive water chemistry of the coolant is maintained [Verga et al., 2001].

Corrosion products as forms of metal oxides with radioactivity in the primary systems accumulate on the surface of equipment and components of nuclear power plants, thus increasing the radiation dose. Various approaches are available for radiation dose management in nuclear power pants in order to reduce the probability of radiation exposure to workers [Yim and Ocken, 2001]. Decontamination is generally accepted as an important means to eliminate or to lower radiation level of radioactive contaminants existing on the surfaces of buildings, equipment, components, system, etc., which are excessively exposed to radiation in nuclear power plants [Ocken, 1999; Wood and Spalaris, 1989; Varrin, 1996; Moon et al., 1997].

The decontamination methods can be classified into mechanical, electrical, and chemical methods. Of decontamination methods, chemical decontamination is considered to be an effective method to dissolve radioactive corrosion products and metal oxidizing films on the surface of tubes or systems by using a chemical decontamination agent which consists of chemical compounds, such as a chelating agent, a reductant, and an oxidant [Ocken, 1999; Ayres, 1970; Lee et al., 2004]. It is of importance to determine chemical species and compositions in a chemical decontaminating agent according to metal species and composition of dissolved metal oxides for the development of a chemical decontamination process. In a previous study, a chemical decontamination process was developed by the dissolution experiments of simulated metal oxides having similar metal species and composition with metal oxides existing in the primary systems of a Korean nuclear power plant [Kim et al., 2003a].

The developed chemical decontamination process consists of four consecutive steps, such as oxidation, reduction, dissolution, and decomposition/cleanup steps. In order to increase the dissolution effectiveness of metal oxides, especially having 10% of chromium or more, the oxidant mixture of $KMnO_4$ and H_3PO_4 is used in the oxidation step as the first step of the chemical decontamination process. In this step, chromium (+3) in the oxide film is oxidized to the more soluble chromate (+6) in a dilute solution of $KMnO_4$ and H_3PO_4. Then, MnO_2 and $HMnO_2$ produced from the oxidation step are reduced in the reduction step. Furthermore, a chemical decontaminating agent is supplied in the dissolution step so as to dissolve metal oxides. This step is known to be one of the critical steps to determine the efficiency of the decontamination process [Ocken,

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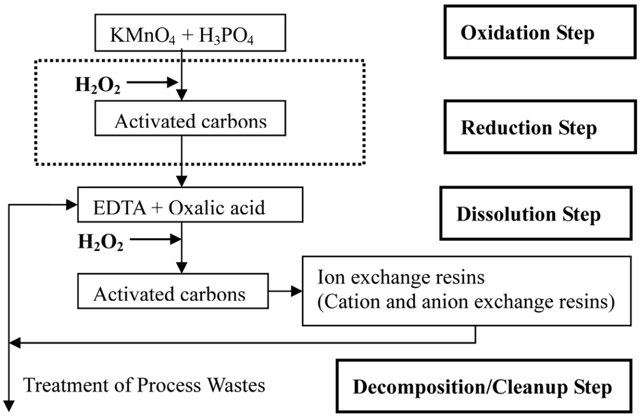
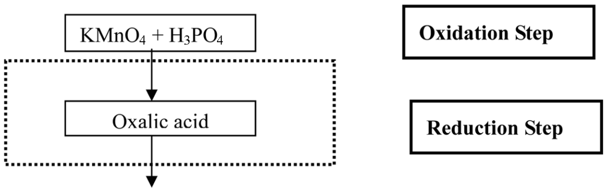


(a) Method 1: adsorption on the adsorbent surface in the reduction step



(b) Method 2: chemical reduction with a reductant in the reduction step

Fig. 1. Different reduction methods in the chemical decontamination processes.

1999]. During the decomposition/cleanup step of the chemical decontamination process, dissolved metal species and their complexes of organic compounds are degraded in the system of hydrogen peroxide and activated carbon. Since activated carbon is a carbonaceous material having a high porosity and a high surface area, it has been extensively used as catalyst or catalyst support as well as adsorbents in a variety of industrial and environmental applications (i.e., the purification and chemical recovery operations as well as removal of organic compounds and metal species) [Kim et al., 1985, 2002, 2003b; Lücking et al., 1998]. Remaining metal species, organic compounds, and metal complexes of organics can be removed by using cation and anion exchange resins.

Two different reduction steps of the chemical decontamination process were considered in this study for the operation of semi-pilot scale decontamination equipment as a continuous work [Kim et al., 2004]. For Method 1, an adsorbent (activated carbon) with an oxygen source (H_2O_2) is used in the reduction step as shown in Fig. 1(a). It is assumed that metal species produced from the oxidant step are removed mainly by adsorption on the surface of activated carbons [Faur-Brasquet et al., 1996; Dastgheib and Rockstraw, 2001]. Oxalic acid is used in the reduction step instead of an adsorbent and an oxygen source for Method 2 (See Fig. 1(b)). Oxalic acid is commonly used as a chelating and/or a reducing agent in many chemical decontamination processes since it is a chemical compound having two carboxyl groups directly joined [Ocken, 1999]. In the present study, the influence of two different reduction steps on the dissolution and removal of metal species and organic compounds was investigated through the developed chemical decontamination process operation. Also, the effects of metal species and organics adsorption on the properties of the adsorbent were investigated through nitrogen adsorption experiments.

EXPERIMENTAL

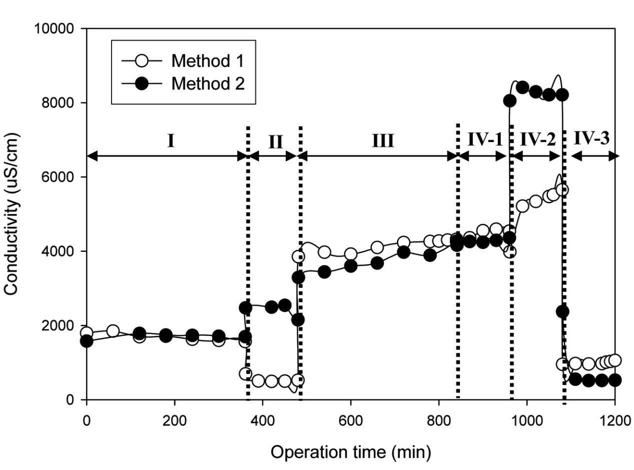
The semi-pilot scale decontamination equipment with a process volume of 120 L was operated with the experimental conditions as shown in Table 1. Simulated metal oxide was prepared by a synthetic method (CRUD 10) using oxide compounds (Fe_3O_4, NiO, and CrO_3) as described in the previous study [Kim et al., 2003a]. Chemical species and their concentrations for each step were determined in terms of the dissolved metal concentration and the dissolution of simulated metal oxides. Detailed specifications of the equipment are found elsewhere [Kim et al., 2004]. The operation temperature was set as 85 °C for each step during the operation of the developed chemical decontamination process. All the chemicals for the experiments were obtained from Junsei Chemical, Japan. Ion exchange resins, DOWEX 650 C and 550 A, were purchased

Table 1. Experimental conditions for two chemical decontamination processes

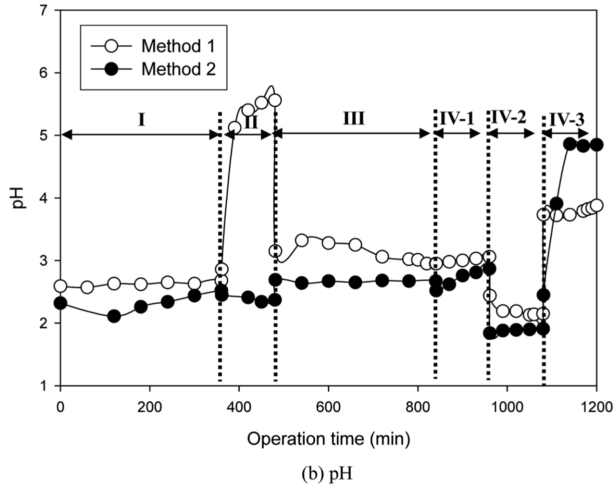
Process step		Experimental conditions			
		Method 1		Method 2	
Oxidation (I) (6 hours)		$KmnO_4$ (0.05%) H_3PO_4 (0.025%) Metal oxide	70.2 g 63 mL 30.3 g	$KMnO_4$ (0.05%) H_3PO_4 (0.025%) Metal oxide	70.5 g 62 mL 30.1 g
Reduction (II) (2 hours)		Activated carbon H_2O_2 (0.03%)	5 L 70 mL	Oxalic acid (0.2%)	281.3 g
Dissolution (III) (6 hours)		Na_2EDTA (0.5%) Oxalic acid (0.2%)	702.2 g 284.6 g	Na_2EDTA (0.5%) Oxalic acid (0.2%)	700.5 g 282.5 g
Decomposition/ Cleanup (IV) (9 hours)	Decomposition (IV-1) (3 hours)	Activated carbon H_2O_2 (0.03%)	5 L 71 mL	Activated carbon H_2O_2 (0.03%)	5 L 70 mL
	Cation exchange (IV-2) (3 hours)	DOWEX 650C	5 L	DOWEX 650C	5 L
	Anion exchange (IV-3) (3 hours)	DOWEX 550A	5 L	DOWEX 550A	5 L

from Dow Chemical, USA as forms of H^+ and OH^-, respectively. Activated carbon having the size of 8-32 mesh (industrial grade) was used as an adsorbent in a 10 L of an activated carbon tower for the reduction and the decomposition/cleanup steps. Oxalic acid was used in the reduction step for the decontamination process of Method 2. The dissolution efficiency of metal species increased with increasing oxalic acid concentration (up to 0.3%). In a previous study, however, oxalic acid concentration for the chemical decontamination process was determined as 0.2% since it can cause an intergranular attack (IGA) on base materials [Kim et al., 2004].

The properties of activated carbon were characterized from the nitrogen adsorption-desorption at 77 K using Micrometrics ASAP 2010 [Toles et al., 1997]. Concentration of organic compounds was analyzed in terms of total organic carbon using a total organic carbon analyzer, Multi N/C 3000 (Analytik Jena AG, Germany) after filtration with 0.45 μm membrane filters. Concentrations of metal species were analyzed by using an inductively coupled plasma-atomic emission spectroscopy (ICP-AES), Optima 4300 DV, Perkin Elmer. In addition, deposition of metal species on the adsorbent surface was analyzed by using an energy dispersive X-ray (EDX) analytical spectrometer (Kevex Sigma Gold, USA) connected to a scanning electron microscope (SEM) (S-4700, Hitachi, Japan).



(a) Conductivity



(b) pH

Fig. 2. Conductivity and pH changes during decontamination experiments (description of steps appeared in Table 1).

RESULTS AND DISCUSSION

1. Observation of Experimental Parameters for Two Decontamination Processes

Four consecutive steps (oxidation, reduction, dissolution, and decomposition/cleanup steps) were operated by using the semi-pilot scale decontamination equipment and the operating parameters were measured for different decontamination processes (Methods 1 and 2). Fig. 2 shows changes in conductivity and pH values according to the elapsed time during the chemical decontamination experiments. When the conductivity change is considered in Fig. 2(a), the results of all steps for two processes are similar except for the reduction step (II) and the cation exchange step (IV-2). Method 1 shows lower conductivity values than Method 2 due to the removal of dissolved metals by adsorption on the surface of activated carbon in the reduction step. The higher conductivity values of Method 2 in the reduction step are assumed to be due to the formation of the metal complex with a reductant (oxalic acid). In addition, higher values of the conductivity during the cation exchange step imply that metal complexes of organic compounds (oxalic acid and EDTA (Ethylenediaminetetraacetic acid)) were not nearly removed because of their negative charges.

Fig. 2(b) shows the pH changes during the operation of two decontamination processes. Like the conductivity change, different results for the reduction and decomposition/cleanup steps between two processes were observed in the pH changes. The lower pH values in the reduction step for Method 2 are due to addition of oxalic acid. In addition, the pH values of the process solution for Method 2 represented higher than Method 1 in the anion exchange step (IV-3). The result implies that metal species were more easily removed during the operation of Method 2 by adsorption on the anion exchange resins as a form of their complex with organic compounds [Song et al., 2005].

2. Dissolution and Removal of Metal Species in Decontamination Processes

The results of metal species concentration changes during the decontamination experiments are shown through Figs. 3 and 5. Fig. 3 shows Fe concentration change for two decontamination pro-

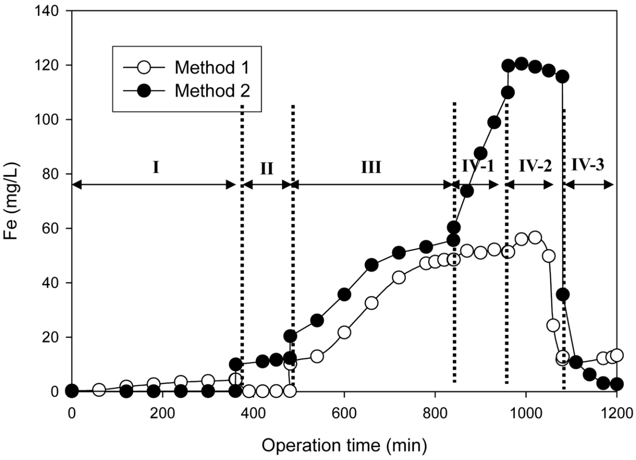


Fig. 3. Changes of Fe concentrations during decontamination experiments.

cesses. For the operation of Method 2, addition of oxalic acid increased the solubility of Fe during the reduction step (II) and the following dissolution step (III). Meanwhile, Fe nearly did not dissolve during the reduction step (II) for Method 1 where metal species were removed by adsorption on the surface of activated carbons. The concentration of Fe shows higher value during the cation exchange step (IV-2), implying that Fe exists as a form of negatively charged oxalate complex rather than positively charged metal species itself. It is assumed that the dissolved Fe was removed as forms of negatively charged organics complexes (Fe : EDTA and Fe : Oxalate) by anion exchange resins in the step of IV-3.

As mentioned before, Cr is extremely insoluble since it exists in the inner oxide layer next to the bulk metal as a form of Fe-chromite. Therefore, increase in the dissolution of Cr is one of the most important considerations for the development of a chemical decontamination process [Verga et al., 2001]. Fig. 4 shows the concentration changes of Cr according to the elapsed time of decontamination experiments. Unlike dissolution of Fe, the dissolved Cr concentration increased notedly in the reduction step for Method 2. It is assumed that the Cr dissolution increased by complexation of oxalate and chromate (+4) in the reduction step after chromium (+3) was oxidized to the more soluble chromate (+6) in the oxidation step [Faur-Brasquet et al., 1996]. The removal of Cr in the final step (IV-3) for Method 2 is related to removal of negatively charged organics complexes of Cr.

Fig. 5 shows change in the dissolved concentration of Ni for two decontamination processes. For the decontamination process of Method 1, the resulting concentration values were very low in the oxidation and reduction steps and then the metal dissolution increased during the dissolution step. A significant increase in the dissolution

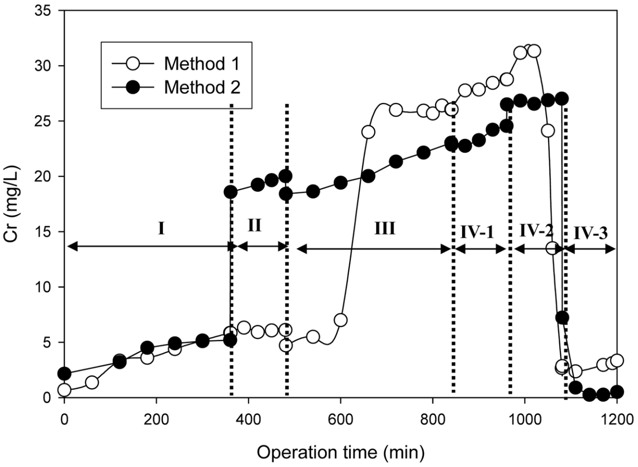


Fig. 4. Changes of Cr concentrations during decontamination experiments.

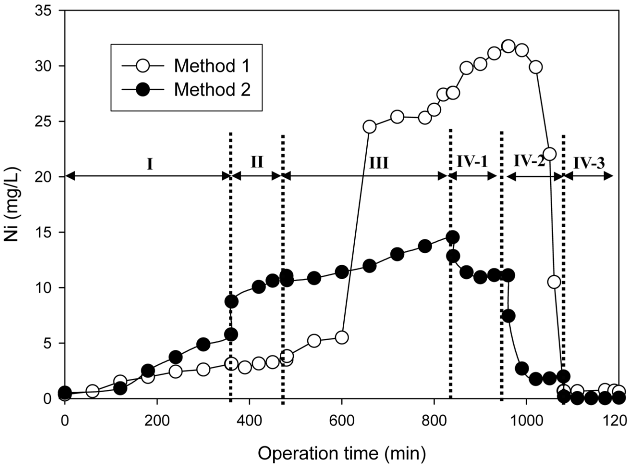


Fig. 5. Changes of Ni concentrations during decontamination experiments.

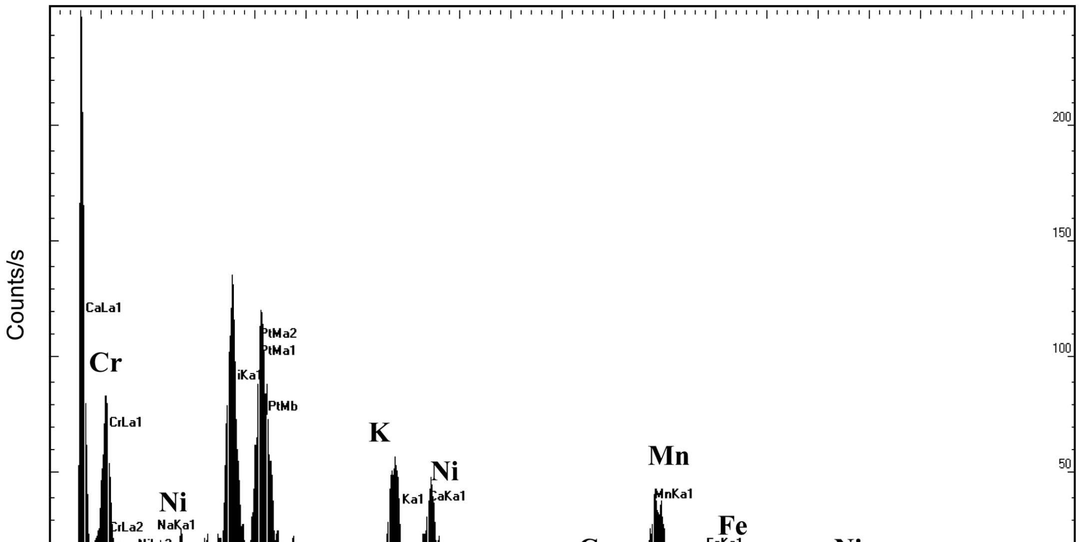


Fig. 6. SEM-EDX image showing adsorption of metal species on the activated carbon surface.

of Ni during the dissolution step is related to formation of complex with EDTA, one of the chemical species in the decontaminating agent [Juang and Wang, 2003]. The concentration of Ni at the final of the dissolution step in Method 1 increased up to 31.76 mg/L. For Method 2, the dissolved Ni concentration in the reduction step was higher compared with the result of Method 1, assuming that the dissolution occurred mainly due to the formation of oxalate complex of Ni. The removal efficiencies of Ni at the final anion exchange step were similar for two decontamination processes. It is noted that addition of oxalic acid in the reduction step increased dissolution by formation of oxalate complex in the results of the dissolution of metal species (Fe, Cr, and Ni). Also, it is thought that dissolved metal species were removed by adsorption on the surface of activated carbon. Adsorption of metal species (Fe, Cr, Ni, K, and Mn) on the activated carbon surface was analyzed by using SEM-EDX and the result is shown in Fig. 6.

In order to investigate the contribution of the reduction step for the dissolution of metal oxide, the dissolved concentration of metal species in the reduction (II) and the dissolution step (III) were compared for two decontamination processes and the results are shown in Table 2. The dissolved concentrations of Fe, Cr, and Ni for Method 2 were much higher compared with the results of Method 1. The dissolved concentration of Fe was 0.2 mg/L for Method 1, while Method 2 showed a concentration of 12.3 mg/L. Considering the ratio of the concentration in step II to III, Method 1 showed 0.4%, 23.5%, 12.7% for Fe, Cr, and Ni, respectively. In the case of Method 2, the contribution of reduction step increased significantly, showing an estimated value of 22.1% for Fe, 86.9% for Cr, and 76.6% for Ni. It is noted that the reduction step using oxalic acid increased the dissolution of Cr for the decontamination process of Method 2.

Table 3 summarizes the results for two decontamination processes in terms of the dissolution of metal species and their removal efficiencies. The concentration of dissolved Cr was similar for two decontamination processes. In the case of the removal efficiency of Cr, Method 2 showed much higher value (98.1%) than Method 1. For the dissolution of Ni, however, the dissolved concentration in Method 1 was higher than that in Method 2 due to the complexation of EDTA. It is noted that the dissolved concentration of Fe was 120.5 mg/L in Method 2, a much higher value than Method 1 (56.6 mg/L). The removal efficiencies of metals in Method 2 showed 97% or above except for K as shown in the table. However, the removal efficiencies of Fe, Cr and K were lower than 90% in Method 1. The experimental results imply that chemical reduction is more effective in the decontamination process than the physical adsorption for removals of metal species.

Table 2. Influence of reduction method on the dissolution of metal oxide for two decontamination processes

		Fe	Cr	Ni
Method 1	Reduction step (C_R) (mg/L)	0.2	6.1	3.5
	Dissolution step (C_D) (mg/L)	48.5	26.0	27.6
	100×(C_R/C_D) (%)	0.4	23.5	12.7
Method 2	Reduction step (C_R) (mg/L)	12.3	20.0	11.1
	Dissolution step (C_D) (mg/L)	55.6	23.0	14.5
	100×(C_R/C_D) (%)	22.1	86.9	76.6

Table 3. Dissolution and removal of metal species for two decontamination processes

	Method 1		Method 2	
	Dissolved concentration (mg/L)	Total removal efficiency (%)	Dissolved concentration (mg/L)	Total removal efficiency (%)
Cr	31.3	89.6	27.0	98.1
Ni	31.8	97.8	14.5	99.5
Fe	56.6	76.1	120.5	97.7
K	146.2	80.2	244.6	92.3
Mn	197.7	99.8	227.2	99.8

3. Dissolution and Removal of Organic Compounds in Decontamination Processes

Of organic compounds existing in the chemical decontaminating agent, EDTA is commonly used as a chelating agent in order to remove metal species in many areas. The completely deprotonated (or dissociated) EDTA molecule is capable by donation of six separate unpaired electrons on the dissociated molecule to complex with one-to-one stoichiometry [Nowack and Sigg, 1996; Seco et al., 1997]. It is known that EDTA has a low rate of biodegradation especially after the formation of complex with metal species [Nowack et al., 1996]. Therefore, chemical oxidation methods are commonly considered for its degradation. Interest in advanced oxidation process with oxidants has increased due to a high reaction rate and a target non-specific nature of radicals among chemical oxidation methods [Ridge and Sedlak, 2004; Frim et al., 2003]. Hydrogen peroxide is considered as one of the effective chemical oxidants since it can produce a highly reactive radical (OH·) in the presence of iron salts or supported iron oxides [Kong et al., 2003; Choi et al., 2001]. In addition, reaction rate increases and the degradation is enhanced on the surface of adsorbents (i.e., activated carbon) having large microporous structures [Ravikmuar and Gurol, 1994; Lee et al., 2003; Khalili et al., 2000].

Fig. 7 shows the results of the removal of organic compounds for two decontamination processes. Considering the change in organics concentration in the figure, two processes showed similar results in the respective dissolution step. During the decomposition/cleanup

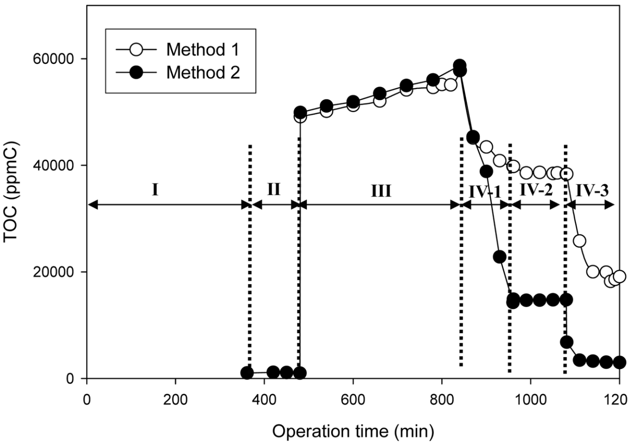


Fig. 7. Changes of organics concentrations during decontamination experiments.

Table 4. Dissolution and removal of organic compounds for two decontamination processes

Process step		Method 1	Method 2
Decomposition step (IV-1)	Concentration at initial (ppmC)	57,772	59,500
	Concentration at final (ppmC)	39,776	14,874
	Removal efficiency in this step (%)	31.2	75.0
Anion exchange step (IV-3)	Concentration at initial (ppmC)	38,357	14,771
	Concentration at final (ppmC)	19,107	2,985
	Removal efficiency in this step (%)	50.2	79.8
Total removal efficiency (%)		66.9	95.0

step, however, a greater amount of organic compounds was removed for the operation of Method 2. It is suggested that organic complex of metal species was more easily degraded by adsorption and chemical conversion due to radicals in the presence of H_2O_2 and iron on the adsorbent during the decomposition step [Ravikmuar and Gurol, 1994; Lücking et al., 1998]. Table 4 summarizes the removal of organic compounds in the steps of IV-1 and IV-3 as well as the total removal efficiency for two decontamination processes. The total removal efficiency for Method 1 was 66.9%. However, Method 2 showed much higher removal efficiency (95.0%). In addition, Method 1 showed a removal efficiency of 31.2% and 50.2% for the decomposition step (IV-1) and the anion exchange step (IV-3), respectively. The removal efficiencies of the two steps for Method 2 were much higher than those for Method 1, showing values of 75.0% and 79.8%, respectively. Because the removal of the remaining organic compounds is one of the most important considerations after a chemical decontamination process operation, the reduction step using oxalic acid would be one of the important considerations in order to increase the decontamination process efficiency [Ocken, 1999; Kim et al., 2004].

4. Influence of Adsorption on the Adsorbent Properties

Activated carbon was used in this study to remove metal species and organic compounds by adsorption on the adsorbent surface and chemical conversion using an oxygen source (H_2O_2) for two decontamination processes. During the operation of the decontamination process, fresh activated carbon was used in an activated carbon tower for the reduction step (II) and the decomposition step (IV-1), respectively. And the properties of activated carbon used in the respective step were compared with those of fresh activated carbon with the adsorption-desorption isotherm as shown in Fig. 8. The isotherm curves in the figure resemble Type I according to the BDDT (Brunauer, Deming, Deming and Teller) theory [Gregg and Sing, 1982]. It is known that the Type I adsorbent has mainly microporous structures and a relatively small external surface area [Khalili et al., 2000; Warhurst et al., 1997]. As shown in the figure, the aspects of adsorption-desorption were similar between the fresh activated carbon (AC_0) and the activated carbon used in the reduction step (AC_R), implying that metal adsorption on the activated carbon surface did not have an effect on the surface properties. However, a notable difference in the adsorption-desorption was observed for the activated carbon used in the decomposition step (AC_D) compared with the result of AC_0, showing that the adsorption amount on the adsorbent surface was notably decreased.

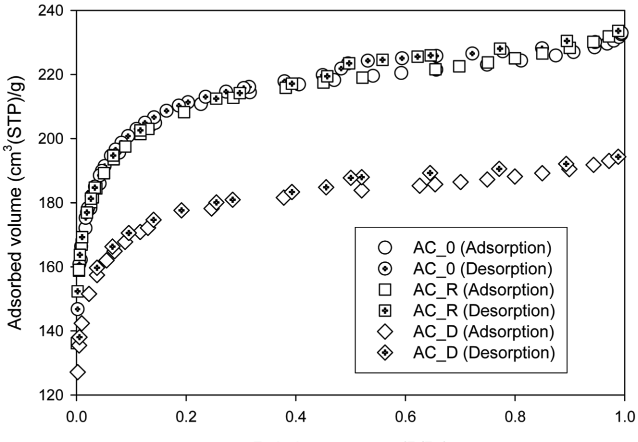


Fig. 8. Influence of metals and metal/organics on the nitrogen adsorption isotherms for activated carbons.

Table 5 summarizes the activated carbon characteristic values (the surface area, pore diameter and the pore volume) for three activated carbons (AC_0, AC_R, and AC_D). Little change was observed in the properties of AC_R compared with those of AC_0 in the table. This result implies that the adsorption of metal species did not affect the properties of the activated carbon during the reduction step (II). However, significant decreases in characteristic values between AC_0 and AC_D were clearly observed due to adsorption of metal species and organic compounds. Adsorption of metal species and organic compounds decreased all values of the properties of the adsorbent. As shown in the table, the micropore surface area and the pore volume mainly decreased to 15% and 16%, respectively, even though the pore diameter for each activated car-

Table 5. Characteristic values for the surface properties of activated carbons

	AC_0[a]	AC_R[b]	AC_D[c]
Area			
BET surface area (m^2/g)	796.74	800.03	678.13
Micropore surface area (m^2/g)	782.33	781.88	665.37
External surface area (m^2/g)	14.41	14.15	12.77
Pore diameter (BET) (Å)	18.09	18.09	18.07
Pore volume (BET) (cm^3/g)	0.33	0.33	0.28

[a] AC_0: fresh activated carbon.

[b] AC_R: activated carbons used in the reduction step (II).

[c] AC_D: activated carbons used in the decomposition step (IV-1).

bon was not changed. It is considered that the properties of the adsorbents were affected more by organic adsorption rather than metal species adsorption.

CONCLUSIONS

In this study, the process efficiencies of two chemical decontamination processes having different reduction steps with physical adsorption (Method 1) and chemical reduction (Method 2) were investigated by the operation of the semi-pilot scale decontamination equipment. The dissolution efficiencies of metal species in Method 2 were higher than in Method 1 due to the formation of metal complexes with excess of oxalic acid. Of metal species, the contribution of the reduction step on the total Cr dissolution showed a significant high value (86.9%). The higher removal efficiency for Method 2 can be explained by the removal of oxalate complex by using chemical conversion in the system of an oxygen source and an adsorbent. It is noted that the removal efficiency of organic compounds, which is one of the important consideration for the development of decontamination process, was 95.0% for Method 2, much higher than Method 1 (66.9%). Through operation of two decontamination processes with different reduction step, it was shown that the process employing chemical reduction showed higher dissolution and removal efficiencies rather than the process by the physical adsorption on the adsorbent surface.

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