



## Original Article

## Performance evaluation of TEDA impregnated activated carbon under long term operation simulated NPP operating condition

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## ABSTRACT

The methyl iodide (CH<sub>3</sub>I) removal performance of tri-ethylene-di-amine impregnated activated carbon (TEDA-AC) used in the air cleaning unit of nuclear power plants (NPPs) should be maintained at least 99% between 24 month-performance test period. In order for evaluating the effectiveness of TEDA-AC on the removal performance of CH<sub>3</sub>I in nuclear power plant during the operation of NPPs, the long-term test for up to 15 months was carried out under the simulated operating conditions (e.g., 25 °C, RH 50%, ppb level poisoning gases injection) at nuclear power plants (NPPs). The TEDA-AC samples were analyzed with the Brunauer-Emmett-Teller (BET) specific surface area and TEDA content as well as CH<sub>3</sub>I penetration test. It is clearly evident that more than 99% of CH<sub>3</sub>I removal performance of TEDA-AC was observed in the TEDA-AC samples during 15 months of long-term operation under the simulated NPP operating conditions including the ppb level of organic and oxide form of poisoning gases. BET specific surface area and TEDA content that can affect the CH<sub>3</sub>I removal performance of TEDA-AC were also maintained as those in new TEDA-AC during 15 months of long-term operation.

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## 1. Introduction

An air cleaning unit (ACU) in the NPPs is used for indoor air quality improvement. The ACU installed at NPPs is the engineered safety feature which is devised to not only reduce the release of the radioactive material to the environment, but also maintain working condition for operators in the main control room. The particulate iodine in the radioactive gases is removed from the high-efficiency particulate air (HEPA) filter installed in the ACU, and the gaseous iodine (elemental iodine and organic iodide) is removed with TEDA-AC in the ACU adsorber. The degradation of impregnated activated carbon is largely due to an aging, weathering, and poisoning. The aging and weathering are caused by the oxidation of activated carbon surface where the oxygen group on activated carbon surface is converted into CO or CO<sub>2</sub>, desorption of impure active substance and adsorption of various substances coexisting in inflow air [1,2]. The TEDA-AC stored in a drum may progress aging gradually even though it is not exposed to outside air at all. Eventually, the organic iodide such as methyl iodide (CH<sub>3</sub>I) removal performance would be gradually decreased. On the other hands,

the poisoning is the exposure of activated carbon to outside air containing oxygen, moisture, various types of pollutants (such as a nitrogen and sulfur oxides, low concentrations of pollutants from plant material and equipment) or the adsorption and reaction with solvents, hydrocarbons, and various chemicals. And then, the iodine removal performance of activated carbon is reduced temporarily or permanently. Several studies have been performed for evaluating the life-time of the activated carbon under various temperature/humidity condition. Deitz conducted experiments to evaluate the effect of relative humidity and pH to aging of the activated carbon [3]. Deuber investigated the aging effect of the activated carbon under various temperature/humidity conditions [4]. However, no quantitative study has been performed under the condition of various poisoning gases for over the one year to understand the aging mechanism of the activated carbon.

The methyl iodide penetration test was performed regularly for the safe use of the TEDA-AC in the ACU of the NPP [5]. If the performance is not maintained, it will be replaced. Since the aging, weathering and poisoning processes on TEDA-AC are occurred simultaneously, it is not easy to investigate each effect, independently. The best way to investigate the performance of TEDA-AC is to secure the TEDA-AC sample for methyl iodide penetration test from the ACU installed in the NPP. However, this is not possible

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during the normal operation of the NPP.

In this study, a semi-plant scale test facility, which simulates the same configuration of components in ACU with the same linear velocity and residence time, has been developed to secure the TEDA-AC samples under the simulated operating conditions, such as temperature, relative humidity and poisoning gases, in the NPPs. The TEDA-AC samples periodically taken from the semi-plant scale test facility have been analyzed to confirm the change of its performance during the long term operation by measuring the BET specific surface area, TEDA content, and methyl iodide penetration test.

## 2. Experimental approaches

### 2.1. Semi-plant scale test facility

The semi-plant scale test facility was developed to simulate the ACU and equipped with the same configuration and components of ACU used in NPPs. As shown in Fig. 1, the semi-plant scale test facility consists of three test apparatuses and each of those is serially configured with a flow strainer, a demister, an air heater, a pre-filter, a HEPA filter and a Type III adsorber. The fan was installed at the downstream of the test facility to suck and form the air flows inside the test facility. The one HEPA size square duct, whose dimension is 610 x 610 x 292 mm (width x height x depth), continued from inlet to HEPA filter to meet 0.2 m/s of linear velocity corresponding to 0.5s of residence time. As shown in Fig. 2, the type III adsorber located after the HEPA filter was designed to have four of four inch beds, which is filled with the TEDA-AC [6]. Twelve canisters (50 mm inner diameter and 100 mm length) were vertically installed at the front and rear of the adsorber to secure the TEDA-AC samples. The size of Type III adsorber was larger than the size of HEPA filter to install 12 canisters and was designed with Eq. (1) [6] to meet 0.2 m/s of linear velocity and 0.5s of residence time passing through the four inch bed.

$$T = t(A - B) / K \cdot Q_{bed} \quad (\text{Eq. 1})$$

Where,

- $T$  = Residence time, sec
- $t$  = Thickness of adsorption bed, m
- $A$  = Gross sheet area of all sheets on inlet side or outlet side, whichever is smaller,  $\text{cm}^2$
- $B$  = Total area of baffles, blanks and margins of all sheet,  $\text{cm}^2$
- $K$  = Convert constant ( $1/36 \times 10^3 \text{cm}^3 \text{h} / \text{m}^3 \text{s}$ )
- $Q_{bed}$  = Flow into the beds,  $\text{m}^3/\text{h}$

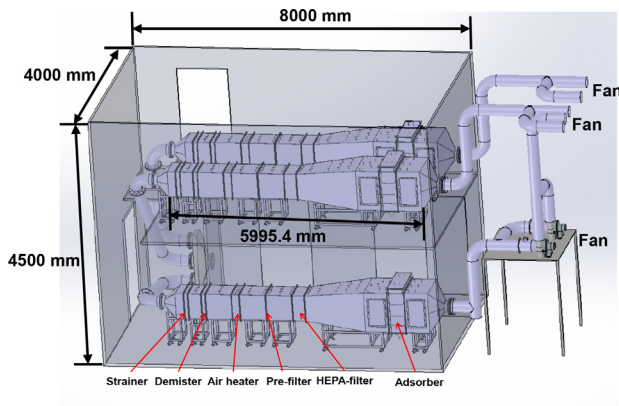


Fig. 1. Schematic drawing of semi-plant scale test facility.

A 90 mm height of reservoir installed above the Type III adsorber (965.2 mm width x 965.2 mm height x 312.4 mm depth) was filled with the TEDA-AC to compensate the volume of TEDA-AC in case of TEDA-AC packing during the operation [6].

The three test apparatuses were installed inside the environmentally well-controlled room to achieve the steady inlet and atmosphere temperatures and humidity, as shown in Fig. 1. Each semi-plant scale test apparatus has different conditions of poisoning gas. Eight different poisoning gases, such as formaldehyde, toluene, acetaldehyde, m-Xylene, p-Xylene, ethyl-benzene,  $\text{SO}_2$  and  $\text{NO}_2$  were supplied into each semi-plant scale test facilities controlled by the regulator and mass flow controller through the gas cylinders.

### 2.2. Test material

The TEDA-AC used in this experiment was a nuclear-grade impregnated activated carbon which is the same lot manufactured by the supplier (Dae-duk.Co.,Ltd) for NPPs. Table 1 shows the physical properties of TEDA-AC used in this experiment, confirming the acceptance criteria required in the ASTM standards. It is seen in Table 1 that the physical properties such as density, particle size distribution and ball-pan hardness meet the ASTM requirements. On the other hand, the initial specific surface area, micro-pore volume and meso-pore volume are  $861.07 \text{m}^2/\text{g}$ ,  $0.36 \text{cm}^3/\text{g}$  and  $0.17 \text{cm}^3/\text{g}$ , respectively, which was analyzed by using well-known analysis methods such as the Brunauer-Emmett-Teller (BET) [7], Horváth-Kawazoe (HK) [8] and Barrett-Joyner-Halenda (BJH) [9]. In addition, 5.057 wt% of the TEDA is initially impregnated on the surface of activated carbon, which was analyzed by the thermogravimetric analysis (TGA).

### 2.3. Test conditions and sampling period

The actual operating conditions of ACU have been inspected in Korean NPPs. The ACU is operating at  $25^\circ\text{C}$  of temperature and 50% of relative humidity. The Type III adsorber in the referenced ACU has 4 inch bed with 0.5s of residence time. As shown in Table 2, three semi-plant scale test apparatuses were operated at the same hydraulic conditions, such as temperature ( $25^\circ\text{C}$ ), relative humidity (50%) and linear velocity (0.2 m/s) corresponding to 0.5 s of residence time to simulate the NPP conditions. On the other hand, the poisoning gas conditions were different for each test apparatus; (1) no poisoning gases in the test apparatus 1, (2) the organic form of poisoning gases such as the formaldehyde, toluene, acetaldehyde, m,p-Xylene and ethyl-benzene in the test apparatus 2, and (3)

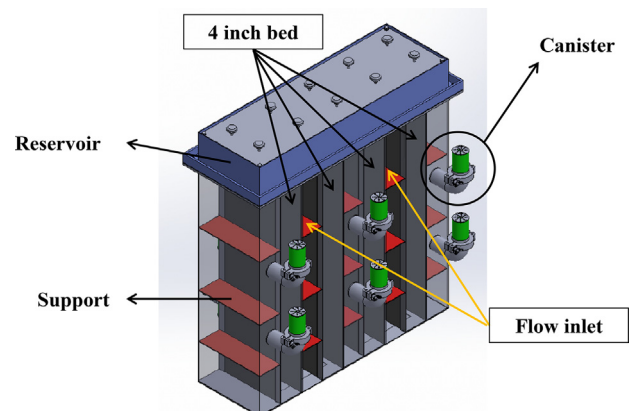


Fig. 2. Schematic drawing of Type-III adsorber.

**Table 1**  
Physical properties of TEDA-AC used in experiment.

Contents		Standard (method)	Acceptance Criteria	Results
Density at dry condition		ASTM D 2854	≥0.38 g/ml	0.54 g/ml
Particle size distribution	Retained on #6 Sieve	ASTM D 2862	≤0.1%	0.0%
	Retained on #8 Sieve		≤5.0%	1.0%
	Through #8, on #12 sieve		≤60%	51.7%
	Through #12, on #16 sieve		≥40%	46.3%
	Through #16		≤5.0%	0.5%
	Through #18		≤1.0%	0.5%
Ball-pan hardness		ASTM D 3802	≥92%	99.7%
Moisture content		ASTM D 2867	State value (%)	3.8%
pH of water extract		ASTM D 3838	State value	10.1
Specific surface area		BET method	–	861.07 m <sup>2</sup> /g
Micro pore volume		HK method	–	0.36 cm <sup>3</sup> /g
Meso pore volume		BJH method	–	0.17 cm <sup>3</sup> /g
Impregnated TEDA Contents		TGA	–	5.057 wt%

the oxide of poisoning gases such as SO<sub>2</sub> and NO<sub>2</sub> as well as the organic poisoning gases in the test apparatus 3. The type and concentration of poisoning gases shown in Table 2 has also been chosen considering the analysis results of air quality samples flowing into the ACU in the NPPs. In order to confirm the change of physical properties such as specific surface area as well as TEDA content in TEDA-AC due to ppb level of poisoning gases. The test conditions for three semi-plant scale test apparatuses are differentiated to see the different poisoning conditions under the same hydraulic conditions.

As described in Section 2.1, the Type III adsorber has twelve canisters for the periodic sampling of the TEDA-AC. Table 3 and Fig. 3 show the identifications (IDs) of canister for the TEDA-AC samples and the sampling period. For example, the canister ID 1 was sampled after one month of operation and then this canister was replaced with new TEDA-AC as the canister ID 13 numbered in the parenthesis which was then sampled after twenty months of operation again. On the other hands, some canisters such as canister ID 1 and 2, 6 and 7, 8 and 9 as well as 10 and 11 were sampled at the same time according to the planned sampling period. The canister ID 16 and 17 replaced after sampling of canister ID 4 and 5 were also sampled at the same time. These two samples were for the confirmation of repeatability of the methyl iodide penetration test. The three test apparatuses had the same IDs of canister and sampling period. The test to secure the periodic TEDA-AC samples continued for twenty four months and finished with The TEDA-AC sample for twenty four months of operation.

### 3. Results and discussions

The long-term operation in the semi-plant scale test facility continued for up to fifteen months under the simulated NPP operating conditions such as 25 °C of temperature, 50% of relative humidity and 0.2 m/s of linear velocity corresponding to 0.5s of residence time through the 10 mm TEDA-AC bed of the Type III adsorber. Fig. 4 shows the inlet thermal-hydraulic conditions during the long-term operation, indicating that the temperature, relative humidity and linear velocity were steady and maintained at 25 ± 1 °C, 50 ± 5% and 0.2 ± 0.018 m/s, respectively during the long term operation up to fifteen months.

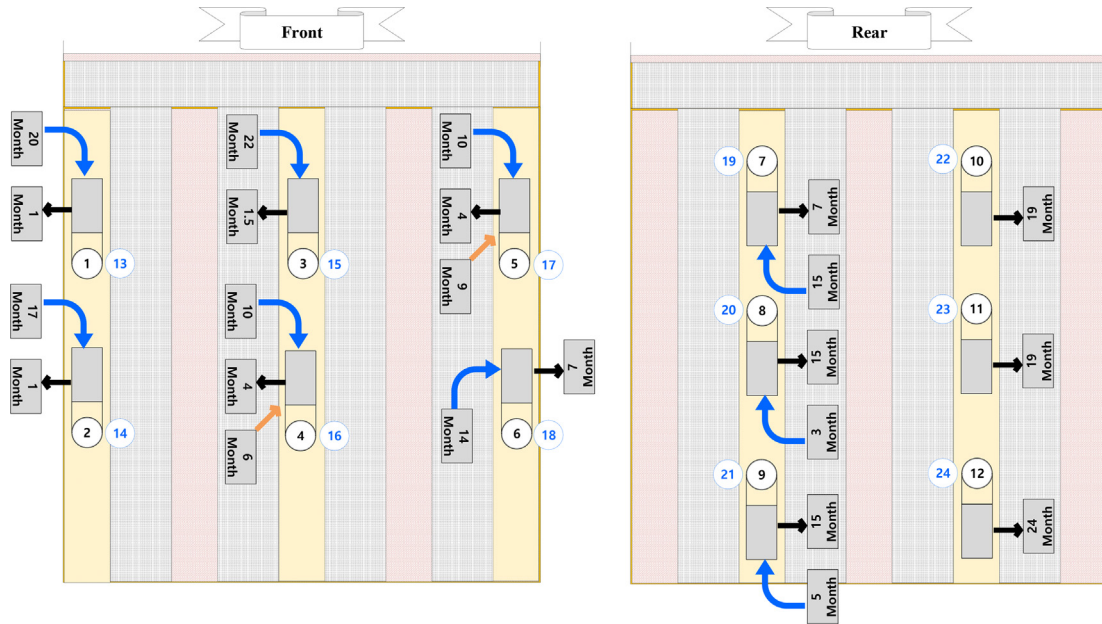
The TEDA-AC samples contained in the canister periodically taken from the Type III adsorber according to the sampling period described in Section 2.3. The samples were analyzed by using the BET specific surface area, TEDA content and methyl iodide penetration test, which have been conducted from the authorized analysis organization in terms of reliability. The surface area of a TEDA-AC can be analyzed by the BET method [7,10]. An adsorption data for nitrogen at the liquid nitrogen temperature, 77K, are usually used in this method. The surface area is taken as the area for monolayer coverage based on the nitrogen molecular area. In the BET method, each site can accommodate 0 to 1 adsorbate molecules, which are not mobile on the surface. On the first layer, the rate of condensation on the bare sites is equal to the rate of evaporation from the sites. Likewise, the equilibrium is established for all layers. Eq. (2) is used to estimate the adsorbed amounts from the BET method as

**Table 2**  
Test conditions of semi-plant scale test apparatus.

Test apparatus	Temperature [°C]	Relative humidity [%]	Linear velocity [m/s]	Residence time [sec]	Poisoning gas	Supplied poisoning gas concentration [ppb]
1	25	50	0.2	0.5	No supply	No supply
2	25	50	0.2	0.5	Formaldehyde	29.28
					Toluene	25.40
					Acetaldehyde m,p-	10.95
					Xylene	22.81
					Ethyl-benzene	11.15
3	25	50	0.2	0.5	Formaldehyde	29.28
					Toluene	25.40
					Acetaldehyde m,p-	10.95
					Xylene	22.81
					Ethyl-benzene	11.15
					SO <sub>2</sub>	10.63
					NO <sub>2</sub>	16.56

**Table 3**  
Canister Identifications (IDs) and corresponding TEDA-AC sampling period.

Canister IDs. (Replaced Canister IDs.)	1	2	3	4	5	6	7	8	9	10	11	12
	(13)	(14)	(15)	(16)	(17)	(18)	(19)	(20)	(21)	(22)	(23)	–
Sampling Period, month	1st	1	1.5	4	4	7	7	15	15	19	19	24
	2nd	(20)	(17)	(22)	(10)	(10)	(14)	(15)	(3)	(5)	–	–
	3rd	–	–	–	(6)	(9)	–	–	–	–	–	–



**Fig. 3.** Canister IDs of TEDA-AC Samples installed in type III adsorber.

$$\frac{V}{V_m} = \frac{cP}{(P_0 - 1) \left[ 1 + (c - 1) \frac{P}{P_0} \right]} \quad (\text{Eq. 2})$$

Where,  
 V = Total adsorption capacity  
 V<sub>m</sub> = Adsorbed amounts  
 P = Pressure  
 P<sub>0</sub> = Saturation pressure  
 c = BET parameter

K = Avogadro's number

σ = Cross-section area (16.2 Å<sup>2</sup> for nitrogen)

The micro-pore volume distribution is analyzed with the Horvath and Kawazoe (HK) method [8]. The HK method is based on slit-shaped pore and basically this method is considered as the molar integral change of the Gibbs free energy. Eq. (4) shows the direct relationship between relative pressure (P/P<sub>0</sub>) and pore size, which is restricted to micro-pore region.

$$RT \ln \left( \frac{p}{p_0} \right) = K \frac{N_a A_a + N_A A_A}{\sigma^4 (l - d)} \times \left[ \frac{\sigma^4}{3 \left( 1 - \frac{d}{2} \right)^3} - \frac{\sigma^{10}}{9 \left( 1 - \frac{d}{2} \right)^9} - \frac{\sigma^4}{3 \left( \frac{d}{2} \right)^3} + \frac{\sigma^{10}}{9 (d/2)^9} \right] \quad (\text{Eq. 4})$$

In order to calculate the surface area, The BET parameter (c) is obtained by the fitting of nitrogen isotherm. The surface area was estimated by Eq. (3) as

$$S_{BET} = V_m \times K \times \sigma \quad (\text{Eq. 3})$$

Where,  
 S<sub>BET</sub> = BET surface area

Where,

N<sub>A</sub> = number of atoms per unit area of adsorbate  
 N<sub>a</sub> = number of atoms per unit area of surface  
 A<sub>a</sub>, A<sub>A</sub> = constant in Lennard-Jones potential  
 l = distance between the nuclei of the two layers  
 σ = distance between a gas atom and the surface at zero interaction energy

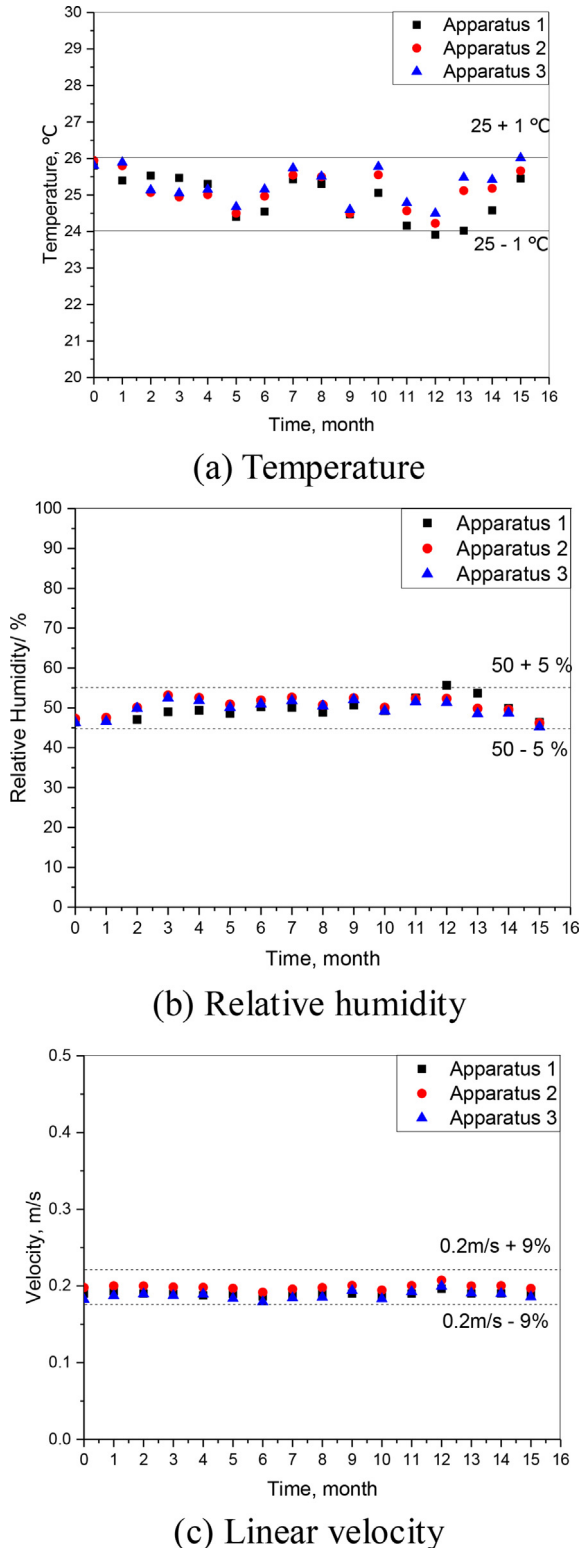


Fig. 4. Inlet thermal-hydraulic conditions during long-term operation of semi-plant test facility.

The Barrett-Joyner-Halenda (BJH) method [9] is used to analyze the meso-pore volume distribution of TEDA-AC. This method is deal with relatively coarse porous adsorbents exhibiting a wide range of pore sizes. The BJH method also uses the adsorption isotherm to analyze the pore size distribution by assuming physical adsorption

and capillary condensation at the pore wall. For this reason, the BJH theory can explain the hysteresis phenomenon in meso-pores of adsorbents.

$$V_s - V = \pi \int_{r_{pn}}^{\infty} (r - t)^2 L(r) dr \quad (\text{Eq. 5})$$

$V_s$  = volume of gas adsorbed at saturation pressure

$V$  = volume of gas adsorbed at pressure,  $p$

$L(r)dr$  = total length of pores whose radii fall between  $r$  and  $r + dr$

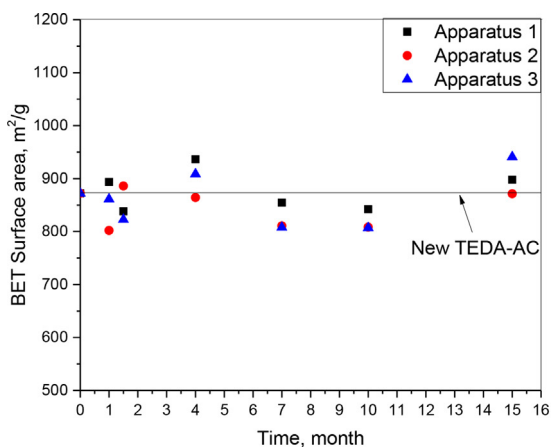
$r_{pn}$  = critical radius.

The specific surface area, micro-pore volume and meso-pore volume of the TEDA-AC samples are shown in Fig. 5.

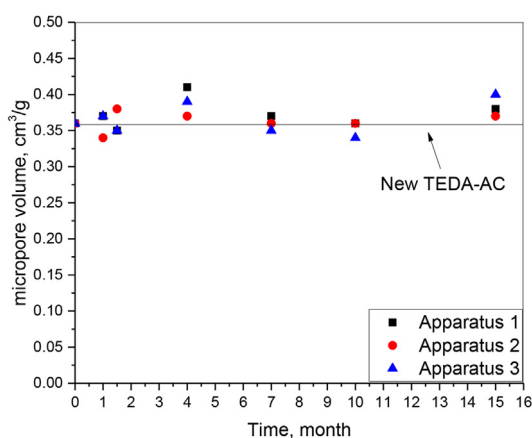
An activated carbon generally has a wide range of pore size distribution containing both micro pore and meso pore. The micro-pore was formed in the internal region and the meso-pore was relatively developed near the surface region of the TEDA-AC [10]. The specific surface area of the activated carbon was reduced by impregnating the TEDA on the surface. Since the micro-pore volume was relatively larger than the meso-pore volume, the micro-pore contributed more to form the specific surface area than the meso-pore. The moisture may adhere to and affect the meso-pore located on the surface of the TEDA-AC during the long term operation. Park et al. investigated the effect of the humidity on the activated carbon that the surface diffusion coefficients are decreased remarkably with the increase of the relative humidity [11].

As shown in Fig. 5, the specific surface area of new TEDA-AC is 861 m<sup>2</sup>/g and those of TEDA-AC samples fluctuate in the vicinity of the specific surface area of new TEDA-AC between 808 m<sup>2</sup>/g and 940 m<sup>2</sup>/g. Since the specific surface area is closely related to the micro-pore volume, the similar behavior with time is observed in these two parameters. Since the thermal-hydraulic conditions such as the temperature, relative humidity and linear velocity are same for three semi-plant scale test apparatuses, the humidity effect on the TEDA-AC expects to be same for three test apparatuses. The differences of specific surface area and meso-pore volume among three test apparatuses may be caused by the difference of poisoning gas conditions. Even though the concentration of poisoning gases is relatively very low, as shown in Table 2, these poisoning gases can be adsorbed physically on the surface of TEDA-AC as well as chemically reacted with TEDA. If some of poisoning gases are physically adsorbed on the surface of TEDA-AC, the surface area would be reduced. It is seen in Fig. 5 (a) that the surface area of test apparatus 1 without poisoning gases is generally larger than that of test apparatus 2 and 3 with an addition of the poisoning gases. Similar behavior is shown in the meso-pore volume in Fig. 5 (c). The meso-pore volumes for the samples taken from the test apparatus 1 were relatively larger than that from the test apparatuses 2 and 3. However, no significant difference on the specific surface areas of TEDA-ACs sampled from the test apparatuses 1, 2 and 3 was observed.

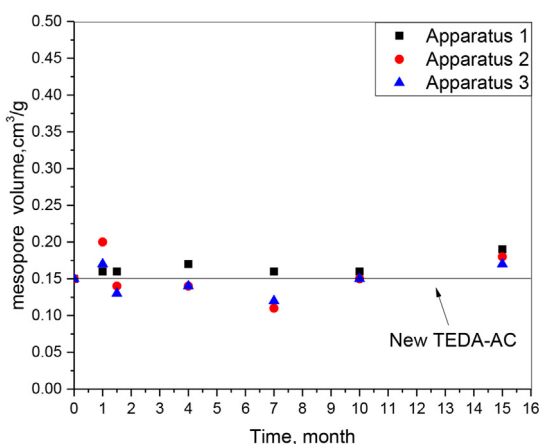
The impregnated activated carbon is used to remove the gaseous iodine, especially organic iodide in relatively high relative humidity conditions. Although the elemental iodine (I<sub>2</sub>) can be captured on the typical activated carbon, the organic iodide is adsorbed on it under the low relative humidity conditions below 30% [12,13]. The impregnation of TEDA is to adsorb the organic iodide efficiently for the high relative humidity conditions. The TEDA reacts with the methyl iodide and then the quaternary



(a) BET specific surface area



(b) Micro-pore volume by HK method



(c) Meso-pore volume by BJH method

Fig. 5. (a) BET specific surface area, (b) micro-pore volume and (c) meso-pore volume of TEDA-AC samples during long term operation.

ammonium salt which is chemically stable is formed [14]. Thus, the TEDA content in the TEDA-AC is closely related to the iodine adsorption performance of the TEDA-AC. Fig. 6 shows the TEDA content analysis for the TEDA-AC samples during the long term operation. The initial TEDA content in new TEDA-AC used for the

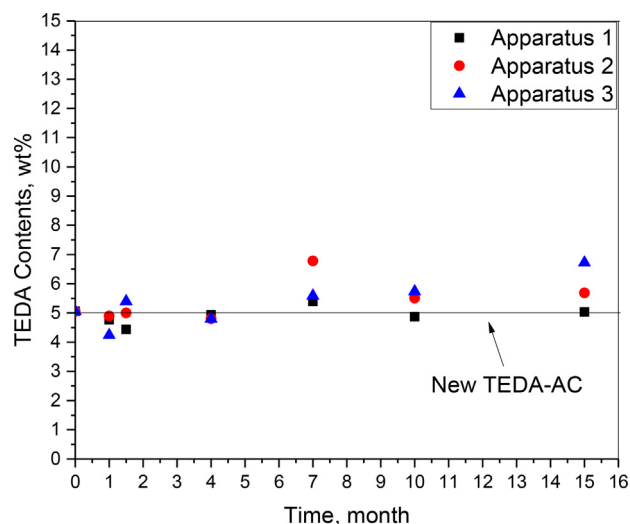


Fig. 6. TEDA content of TEDA-AC samples during long term operation.

NPP is typically about 5 wt% [12].

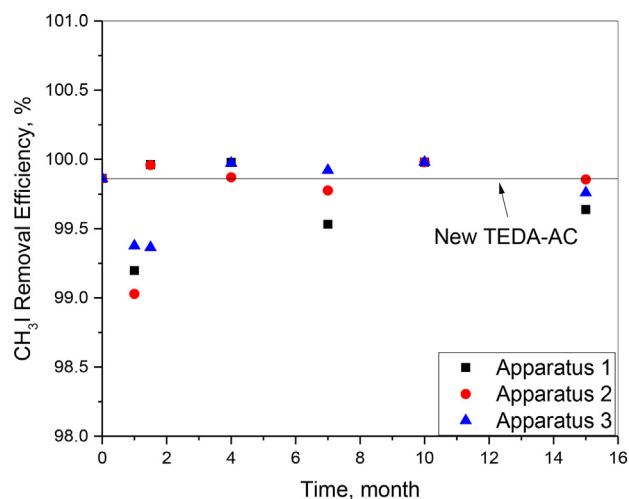
It is seen in Fig. 6 that the TEDA content in new TEDA-AC is about 5 wt% and those in the TEDA-AC samples with time fluctuate in the vicinity of new TEDA-AC between 4.44 wt% and 6.78 wt%. The TEDA contents in TEDA-AC samples after one month and one and half months are slightly reduced from that of TEDA. The TEDA contents in other TEDA-AC samples show the values near or above 5 wt%. Even though the TEDA-AC samples taken from the test apparatus 2 and 3 in the presence of the poisoning gases have relatively higher TEDA content than that taken from the test apparatus 1. The TEDA contents of TEDA-ACs samples did not show the decreasing behavior from the initial TEDA contents during the long term operation for up to fifteen months. Since the TEDA was impregnated on the carbon in bulk during the manufacturing, once may postulate that the initial TEDA content in new TEDA-AC may not be uniformly impregnated on the surface of the activated carbon.

The main role of the TEDA-AC used in the ACU is to adsorb the gaseous iodine. Thus, the performance of TEDA-AC can be determined by the methyl iodide removal capability. The methyl iodide removal performance of the TEDA-AC was analyzed by the methyl iodide penetration test specified in the ASTM D 3803 [5]. The test conditions for the methyl iodide penetration test are shown in Table 4 where the radioactive methyl iodide ( $\text{CH}_3^{31}\text{I}$ ) concentration is  $1.75 \text{ mg/m}^3$  in the flows which corresponds to  $0.2 \text{ m/s}$  of linear velocity under  $30 \text{ }^\circ\text{C}$  of challenge temperature and 95% of relative humidity for 1 h. The test was conducted at authorized test agency for the TEDA-AC samples periodically taken from the semi-plant scale test facility shown in Table 3 and Fig. 3. Fig. 7 shows the methyl iodide removal efficiency for the TEDA-AC samples during the long term operation, indicating the more than 99% removal efficiency. As shown in Fig. 7, the methyl iodide removal efficiencies of TEDA-AC samples taken from the test apparatuses 1, 2 and 3 after one month operation are slightly decreased from that of new TEDA-AC, however all are over 99% and recovered for the subsequent TEDA-AC samples. These slight decreases of methyl iodide removal efficiency of TEDA-AC samples after one month may be caused by the slight decrease of the TEDA contents shown in Fig. 6.

The methyl iodide removal efficiencies for the TEDA-AC samples taken from the test apparatuses 1, 2 and 3 were steady and maintained at above 99.5% for up to fifteen months operation. Even though the poisoning gases were supplied in the test apparatuses 2 and 3, the methyl iodide removal efficiencies were not much different from that of TEDA-AC sample taken from the test

**Table 4**  
Test conditions for methyl iodide penetration test.

Test Parameter	Nominal Condition
Pre-equilibration time (h)	16
Equilibration time (min)	120
Challenge time (min)	60
Elution time (min)	60
CH <sub>3</sub> <sup>131</sup> I concentration (mg/m <sup>3</sup> )	1.75
Test bed depth (mm)	101.6 (4inch)
Test bed diameter (mm)	50
Pre-equilibration temperature (°C)	30
Equilibration temperature (°C)	30
Challenge temperature (°C)	30
Elution temperature (°C)	30
Linear velocity after equilibration (m/s)	0.2
Relative humidity after equilibration (%)	95
Pressure after equilibration (kPa)	101.3



**Fig. 7.** Methyl iodide removal efficiency of TEDA-AC samples during long term operation.

apparatus 1. The low concentrations of poisoning gas concentrations (at ppb levels) supplied in the test apparatus 2 and 3 may not play a significant role and thus more tests at a higher addition of poisoning gases may be needed. On the other hand, the effects of the thermal-hydraulic conditions such as the temperature, relative humidity and linear velocity on the TEDA-AC samples would be also insignificant based on the behavior of the methyl iodide removal efficiencies shown in Fig. 7. It is thought that the performance of TEDA-AC can be maintained during the long term operation up to fifteen months under 25 °C of temperature, 50% of relative humidity and 0.2 m/s of linear velocity which are the same conditions as those in the ACU at NPPs.

#### 4. Conclusion

The performance of TEDA-AC was investigated with the semi-plant scale test facility under the simulated NPP conditions such as at 25 °C, 50% relative humidity, and the linear velocity of 0.2 m/s corresponding to the residence time of 0.5s, with and without an addition of poisoning gases for up to fifteen months. The TEDA-AC used in this test is the same lot of nuclear grade TEDA-AC manufactured for NPPs. The semi-plant scale test facility was developed to simulate the ACU of NPPs. Three test apparatuses installed in the semi-plant scale test facility was designed to have the Type III adsorber with twelve canisters for the TEDA-AC samples. The same thermal-hydraulic conditions, such as the temperature (25 °C),

relative humidity (50%) and linear velocity (0.2 m/s) were used for all three test apparatuses. No poisoning gases were supplied to the test apparatus 1. The poisoning gases such as formaldehyde, toluene, acetaldehyde, m-Xylene, p-Xylene and ethyl-benzene were supplied to the test apparatus 2. On the other hand, the same poisoning gases supplied in the test apparatus 2 as well as SO<sub>2</sub> and NO<sub>2</sub> were applied to the test apparatus 3. During the long term operation for up to fifteen months, the canister installed in the Type III adsorber, which contains the TEDA-ACs were periodically taken for analysis. The TEDA-ACs were independently analyzed by the BET specific surface area with micro and meso-pore volume, TEDA content and methyl iodide penetration test. The following conclusions can be drawn from the test results.

The fluctuation of BET specific surface area and TEDA content for the TEDA-AC samples from new TEDA-AC for up to fifteen months between 808 m<sup>2</sup>/g and 940 m<sup>2</sup>/g and 4.44 wt% to 6.78 wt%, respectively, was observed and the methyl iodide removal efficiencies of the TEDA-AC samples were maintained more than 99% during the long-term operation up to fifteen months. This means that the effectiveness of TEDA-AC could be confirmed to be used in the ACU in NPPs for the long-term up to fifteen months without the consideration of performance degradation by the ppb level of organic and oxide form of poisoning gases. However, the methyl iodide removal performance during the long-term operation up to fifteen months in this study could be confirmed under the operating NPP conditions, e.g., temperature (25 °C), relative humidity (50%), linear velocity (0.2 m/s) corresponding to 0.5 s of residence time and ppb level of poisoning gases including organic and oxide forms.

Even though there were slight decrease of methyl iodide removal efficiency in TEDA-AC samples from that of new TEDA-AC after one month or one and half months operation, the recovery of the methyl iodide removal efficiencies to the level of new TEDA-AC were observed in the subsequent TEDA-AC samples for up to fifteen months. Thus, one can postulate that the performance degradation of TEDA-AC may not be determined with the analysis of TEDA-AC sample under the relatively short operation time.

The test plans to continue further for twenty four months to examine the performance of TEDA-AC.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.net.2020.04.020>.

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