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Evaluation of dose received by workers while repairing a failed spent resin mixture treatment device

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ABSTRACT

Intermediate-level radioactive waste (ILW) is not subject to legal approval for cave disposal in Korea. To solve this problem, a spent resin treatment device that separates ^{14}C-containing resin from zeolite/activated carbon and desorbs ^{14}C through a microwave device has been developed. In this study, we evaluated the radiological safety of the operators performing repair work in the event of a failure in such a device treating 1 ton of spent resin mixture per day. Based on the safety evaluation results, it is possible to formulate a design plan that can ensure the safety of workers while developing a commercialized device. When each component of the resin treatment device can be repaired from the outside, the maximum and minimum allowable repair times are calculated as 263.2 h and 27.7 h for the ^{14}C-detached resin storage tank and zeolite/activated carbon storage tank, respectively. For at least 6 h per quarter, the worker's annual dose limit remains within 50 mSv/year; further, over 5 years, it remained within 100 mSv. At least 6 h of repair time per quarter is considered, under conservative conditions, to verify the radiological safety of the worker during repair work within that time.

1. Introduction

Ion-exchange resins that are used in heavy-water reactors play a significant role in the purification of liquid radioactive waste generated during the operation of a nuclear power plant (NPP) [1]. In an NPP, the desalination tower of the steam generator (SG) purifies water and prevents contamination of the secondary system by removing the radioactive material when the SG tube leaks. An NPP generates ~10,000 L of resin annually. A small amount of radionuclides is detected in this resin owing to the SG tubule microdefects, and this resin is stored in a storage tank in the NPP without treatment/disposal. Therefore, the stored resin must be handled in accordance with future decommissioning plans [2–5].

Radioactive ^{14}C, generated in an NPP, is mainly contained in the moderator (94.5%), nuclear fuel (3.4%), primary heat transport system (1.6%), and annular gas (0.2%) [6]. Double-mixed-phase ion-exchange resins can remove 95% of ^{14}C produced in an NPP. These ^{14}C-containing spent resins are then stored in a resin storage tank. The Wolseong NPP has a resin storage tank capacity of 1786 m^3 (unit 1: 586 m^3, units 2, 3, and 4: 400 m^3 each). When the storage amount is calculated considering the decommissioning time, the maximum amount of resin mixture useable in a heavy-water reactor is found to be 70% of the total storage tank capacity, i.e., 1250 m^3. Storing a corresponding amount of spent resin will require 7813 drums of 200 L capacity with a filling rate of 80%. The cave repository has a ^{14}C total capacity of 1.66 E+14 Bq; however, the total radioactivity of ^{14}C in spent resins at the Wolseong NPP units 1–4 is approximately 1.48 E+16 Bq, a value higher than the total ^{14}C capacity of 1.66 E+14 Bq. Therefore, ^{14}C in the spent resins must be removed. However, the spent resin from heavy-water reactors contains various radionuclides such as ^{3}H, ^{14}C, ^{60}Co, and ^{137}Cs that are classified as intermediate-level radioactive wastes (ILWs) along with a high concentration of ^{14}C, which is a long-lived radionuclide with a half-life of 5730 years [7–10]. According to the radioactive waste classification standards, the ILWs should be disposed of in caves; however, currently, cave disposal of ILW is not allowed in South Korea [11].

To solve this problem, various spent resin treatment devices are currently under development. Such a device can fail at any time under any condition; thus, the safety of the workers, engaged in the repair of such a failed device, should also be ensured. However, radiological safety evaluations for workers repairing various components of such devices have not yet been conducted. In light of this, in this study, the maximum repair time was derived in case of

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component failures when using a commercialized spent resin treatment device to remove ^{14}C from the spent resin as a low-level waste (LLW) for disposal. The results of this study indicate toward a modification in the design of the device as a possible remedy to ensure safety of the repair worker in the case of component failure. The spent resin treatment device can treat maximum 1 ton of spent resin mixture per day; however, it can contain a maximum of 2825 kg of the spent resin mixture. To derive conservative results, the maximum allowable repair time was derived based on the maximum residual amount of the spent resin mixture. The maximum repair time was set based on an 8 h workday, and the maximum number of repair workdays per year was also derived. Assuming that the device could be repaired externally, the maximum allowable repair time was derived considering an external dose. The external dose values were derived using the VISIPLAN 3D as low as reasonably achievable (ALARA) planning tool, which was developed at the SCK-CEN laboratory in Belgium in 1999, for nuclear facilities [12].

2. Methods

2.1. Source information

The activity concentration values of the spent resin mixture, sampled from the spent resin mixture storage tank #2 of the Wolseong NPP unit 1 derived in a previous study, were used in the present work [1]. Based on the activity concentration of the nuclides, radioactivity values were derived using the amount of spent resin mixture included per device (Table 1).

2.2. Modeling of spent resin treatment device

Fig. 1 illustrates the spent resin mixture treatment process [13]. This process involves the following steps: (1) moving the spent resin mixture to a separator; (2) separating the resin from zeolite and activated carbon using the spent resin mixture separator; (3) storing zeolites and activated carbon separately in individual tanks; (4) moving the separated resin to a microwave device; (5) separating ^{14}C using the microwave device; (6) storing the resins separated from ^{14}C in separate storage tanks; and (7) circulating the separated ^{14}C into the adsorption tower and concentrating it in the $CaCO_3$ adsorbent.

If the mixture consists of zeolite, activated carbon, and resin, then it is classified as an ILW due to ^{14}C exceeding the LLW radioactivity criteria attached to the resin. When zeolite and activated carbon are separated from the resin, the waste can be disposed of as an LLW. This implies that ^{14}C is desorbed from the separated waste resin by a spent resin mixture treatment device, and the residual resin can be disposed of as an LLW. The adsorbent is recycled when labeling compounds are manufactured for medicinal purposes. When processing 1 ton of spent resin mixture per day, the maximum processing capacity of the device is 400 kg of spent resin, 100 kg of zeolite, and 100 kg of activated carbon. However, for conservative results, in this study, we derived the maximum allowable repair time based on the fact that the device can contain a maximum of 2825 kg of spent resin mixture, as shown in Table 1.

For the five components of the spent resin treatment device—spent resin mixture separator, zeolite and activated carbon storage tank, resin hopper tank, microwave device, and ^{14}C-detached resin storage tank—the external dose received by workers during repair work was evaluated.

In Fig. 1, lead shielding was assumed for the device to reduce the external dose received by a worker. It was applied to parts of the spent resin mixture separator, zeolite and activated carbon storage tank, resin hopper tank, microwave device, and ^{14}C-detached resin storage tank where the source term was located. Lead shielding was not considered for the adsorption tower. This is because there are beta-emitting nuclides, such as ^{14}C and ^{3}H, inside the adsorption tower, which are protected by the device itself. Based on the ratio of the external dose changes with the thickness of lead shield, derived from existing studies, and the cost aspect of lead, the spent resin treatment device was modeled with a 0.5 cm thick lead shield. When a lead shield of 0.5 cm was applied after no lead shielding, the external dose value decreased by approximately 70.83%. As the lead shielding thickness was increased to 1.0, 1.5, and 2.0 cm, the external dose rate decreased by 83.33%, 90.00%, and 93.89%, respectively [1]. The modeled device was 4 m wide, 1 m long, and 3 m high.

2.2.1. Repairing spent resin mixture separator

The spent resin mixture separator could contain a maximum of 125 kg of the resin mixture. The resin mixture consisted of 10% (12.5 kg) of zeolite, 10% (12.5 kg) of activated carbon, and 80% (100 kg) of resin [1]. Table 1 shows that the maximum radioactivity was 1.73 E+09 Bq. The external dose, during the separator device repair, was evaluated assuming that the repair work was performed in a space 10 cm away from the device. The exposure height was considered at the chest of the worker, that is, 120 cm above ground [14].

2.2.2. Repairing zeolite and activated carbon storage tank

The separated zeolite and activated carbon storage tank could contain a maximum of 1000 kg of zeolite and activated carbon. This mixture consisted of 50% (500 kg) of zeolite and 50% (500 kg) of

Table 1
Radioactivity values when the maximum spent resin mixture for each device is included.

	Spent resin mixture separator (Bq)			Zeolite and activated carbon storage tank (Bq)		Resin hopper tank (Bq)	Microwave device (Bq)	^{14}C detached resin storage tank (Bq)
Nuclide	Zeolite (12.5 kg)	Activated carbon (12.5 kg)	Resin (100 kg)	Zeolite (500 kg)	Activated carbon (500 kg)	Resin (1000 kg)	Resin (200 kg)	Resin (500 kg)
^{57}Co	0.00 E+00	0.00 E+00	2.05 E+06	0.00 E+00	0.00 E+00	2.05 E+07	4.10 E+06	1.03 E+07
^{60}Co	6.23 E+05	1.90 E+06	3.82 E+07	2.49 E+07	7.60 E+07	3.82 E+08	7.64 E+07	1.91 E+08
^{51}Cr	0.00 E+00	0.00 E+00	2.05 E+07	0.00 E+00	0.00 E+00	2.05 E+08	4.10 E+07	1.03 E+08
^{134}Cs	2.99 E+05	2.25 E+04	1.33 E+06	1.20 E+07	9.00 E+05	1.33 E+07	2.66 E+06	6.65 E+06
^{137}Cs	4.03 E+08	2.04 E+07	1.16 E+09	1.61 E+10	8.15 E+08	1.16 E+10	2.32 E+09	5.80 E+09
^{54}Mn	0.00 E+00	0.00 E+00	1.60 E+06	0.00 E+00	0.00 E+00	1.60 E+07	3.20 E+06	8.00 E+06
^{95}Nb	3.61 E+03	7.40 E+04	3.67 E+06	1.45 E+05	2.96 E+06	3.67 E+07	7.34 E+06	1.84 E+07
^{125}Sb	0.00 E+00	1.24 E+05	2.80 E+07	0.00 E+00	4.95 E+06	2.80 E+08	5.60 E+07	1.40 E+08
^{95}Zr	0.00 E+00	0.00 E+00	2.68 E+06	0.00 E+00	0.00 E+00	2.68 E+07	5.36 E+06	1.34 E+07
^{152}Eu	0.00 E+00	0.00 E+00	4.44 E+07	0.00 E+00	0.00 E+00	4.44 E+08	8.88 E+07	2.22 E+08
^{154}Eu	0.00 E+00	0.00 E+00	3.48 E+06	0.00 E+00	0.00 E+00	3.48 E+07	6.96 E+06	1.74 E+07
Total	1.73 E+09			1.70 E+10		1.31 E+10	2.61 E+09	6.53 E+09

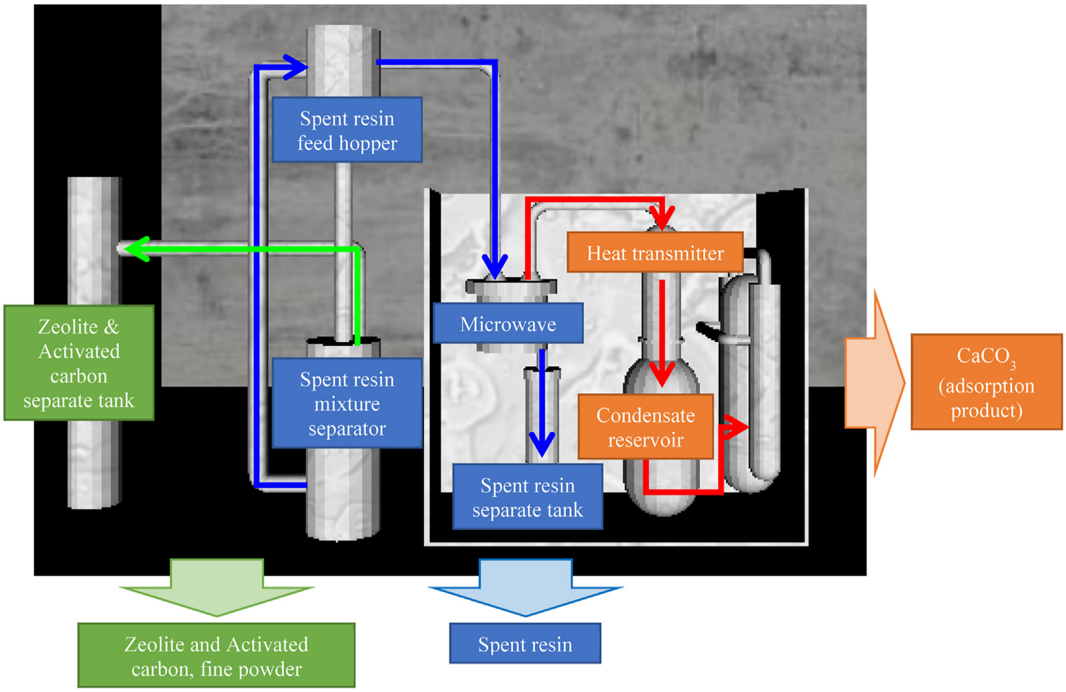

Fig. 1. Spent resin mixture treatment device process.

activated carbon. Table 1 shows that the maximum radioactivity was 1.70 E+10 Bq. When repairing the zeolite and activated carbon storage tank, the external dose was evaluated assuming that the repair work was performed in a space 10 cm away from the device. The exposure height was at the chest of the worker, that is, 120 cm above ground.

2.3. Repairing resin hopper tank

The resin hopper tank stores the resin separated from the spent resin mixture before it is transferred to the microwave device. It can contain a maximum of 1000 kg of resin. Specifically, this tank contained 100% (1000 kg) of pure resin separated from zeolite and activated carbon. Table 1 shows that the maximum radioactivity was 1.31 E+10 Bq. When repairing the resin hopper tank, the external dose was evaluated assuming that the repair work was performed at a distance 10 cm away from the device. The exposure height was 240 cm above the ground considering the location of the resin hopper tank.

2.4. Repairing microwave device

The microwave device, which can contain a maximum of 200 kg of resin, detaches ^{14}C from the resin. Similar to the spent resin hopper tank, it contained 100% (200 kg) of pure resin. Table 1 shows that the maximum radioactivity was 2.61 E+09 Bq. When repairing the microwave device, the external dose was evaluated assuming that the repair work was performed at a distance 10 cm away from the device. The exposure height was at the chest of the worker, that is, 120 cm above ground.

2.4.1. Repairing ^{14}C detached resin storage tank

The ^{14}C detached resin storage tank stores the resin from which ^{14}C is removed using the microwave device. It can contain a maximum of 500 kg of resin. Table 1 shows that the maximum radioactivity was 6.53 E+09 Bq. When repairing the ^{14}C detached resin storage tank, the external dose was evaluated assuming that the repair work was performed at a distance of 10 cm from the device. The exposure height was at the chest of the worker, that is, 120 cm above ground.

2.5. Assessment of external dose from spent resin treatment device during repair

The spent resin mixture treatment device was modeled using VISIPLAN to evaluate the external dose received during the repair work. In the VISIPLAN code, the external dose is calculated using the point kernel method, and the following equations [15]:

$$\phi = \int_V \frac{S \cdot B \cdot e^{-\mu}}{4 \cdot \pi \cdot \rho^2} \tag{1}$$

where.

ϕ : Photon fluence rate $[m^{-2} \cdot {}^{-1}]$.
V: Unit volume $[m^3]$.
S: Source strength per unit volume $[m^{-3} \cdot s^{-1}]$.
B: Build-up factor.
μ: Attenuation effectiveness of a shield.
ρ: Distance from a point source [m].

Each small source is called a kernel, and the integration process in which each point's contribution to the dose is added is called the “point kernel” integration. In our case, the effective dose rate was derived using a dose conversion factor (DCF) based on the photon flux at each point. The DCF in Eq. (2) was derived based on the data provided in ICRP 51 [16].

$$E = \sum_i h_i \Phi_i \tag{2}$$

where.

E : Effective dose rate $[Sv \cdot s^{-1}]$.
h_i: DCF for the photon energy of radionuclide, $i[Sv \cdot m^2]$.
Φ_i: Photon flux for photon energy of radionuclide, $i[m^{-2} \cdot s^{-1}]$.

To solve Eq. (1), a monoenergetic photon source, *S*, is considered. In numerous shielding problems, numerous other sources arise, which emit photons at different energies. The VISIPLAN code uses a formula in which 25 energy bins are used [17]. For calculations using this code, a source spectrum, derived from different calculation codes, is regrouped into 25 energy groups.

2.6. Assessment of internal dose from spent resin treatment device during repair

The internal dose value was so large that the repairable time was close to zero. The largest and smallest internal dose values, across the various components, were derived. Various measures have been proposed to ignore the corresponding internal dose values. For the internal dose assessment, the exposure was assumed to be caused only by inhalation and not by ingestion. Furthermore, the workers wore an air-purifying respirator with an assigned protection factor (APF) of 50 while working [18]. The committed effective dose for 50 years owing to radionuclide inhalation was derived using Eq. (3) [19] shown below:

$$D^{inh} = (\mathrm{BR} \times RC^{inh} \times \mathrm{T}) \times DCF^{inh} \tag{3}$$

where.

D^{inh}: Committed effective dose for 50 years [mSv].

BR: Breathing rate [$\mathrm{m}^3 \cdot \mathrm{h}^{-1}$].

RC^{inh}: Activity concentration [$\mathrm{Bq} \cdot \mathrm{m}^{-3}$].

T: Working time [h].

DCF^{inh}: Dose conversion factor for inhalation [$\mathrm{mSv} \cdot \mathrm{Bq}^{-1}$].

Based on the breathing rate (BR) of adult males during intense work, as specified in ICRP 66, BR in Eq. (3) was set to 1.68 m^3/h [20]. For obtaining the RC^{inh}value, the radioactivity values shown in Table 1 were used, and a value of 1 m^3 value was used for the conservative evaluation of the space where the nuclides are distributed. The value of T was set to 8 h. For the DCF inhalation value, ICRP 119's DCF values were used [21]. For conservative evaluations, results were derived assuming that the committed effective dose for 50 years is received in 1 year.

3. Results and discussions

3.1. Assessment of external dose of spent resin treatment device during repair

When the maximum inclusion capacity for each component of the spent resin treatment device was applied, the distribution of the external dose rate could be confirmed as shown in Fig. 2. A total of 1000 kg of zeolite and activated carbon was distributed in the zeolite and activated carbon storage tank, indicating a high external dose rate around the tank. During the whole repair process, the highest external dose rate (7.23E-01 mSv/h) was obtained during the repair of the zeolite and activated carbon storage tank. In contrast, the lowest external dose rate (7.42E-02 mSv/h) was obtained during the repair of the ^{14}C-detached resin storage tank. The average external dose rate was found to be 3.24E-01 mSv/h during

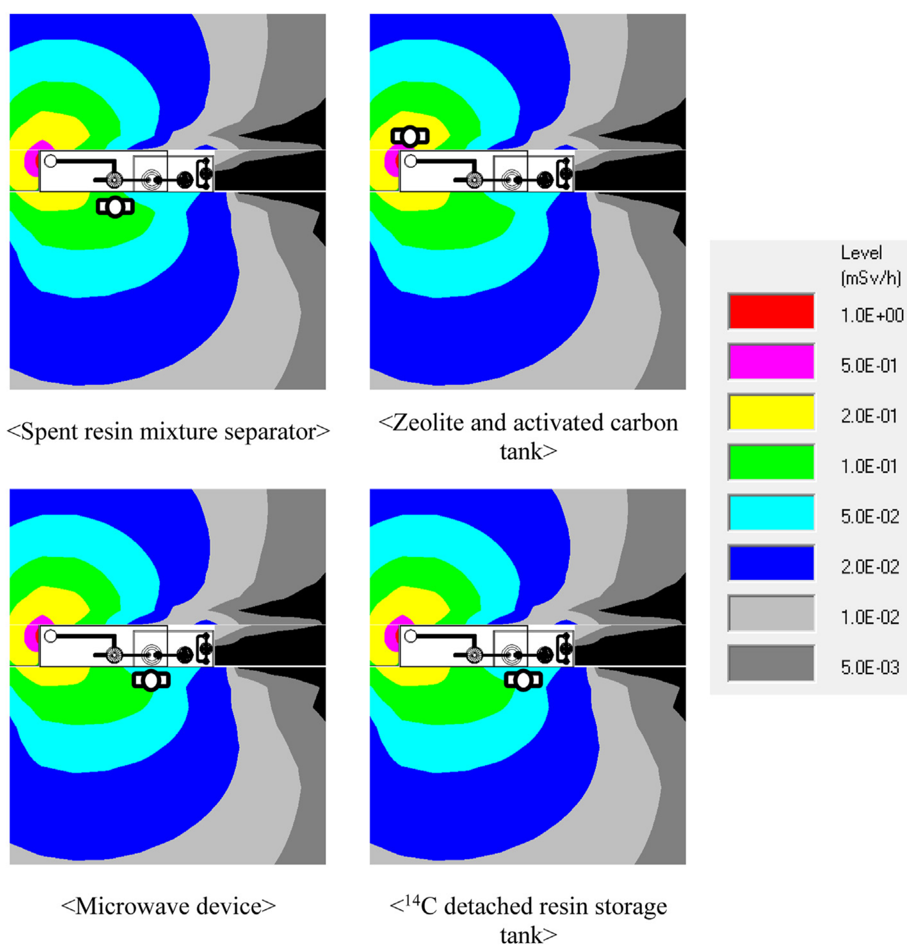

Fig. 2. Distribution of the external dose rate (height is 120 cm).

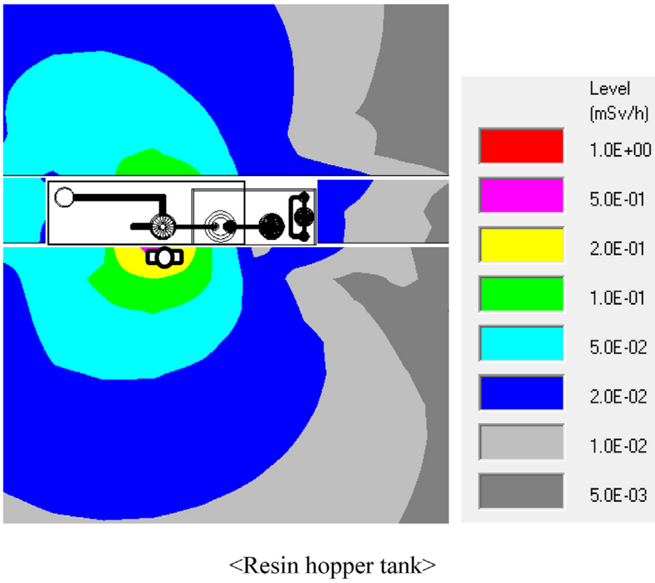

Fig. 3. Distribution of the external dose rate (height is 240 cm).

device repair. Fig. 3 shows the external dose rate distribution obtained while repairing the resin hopper tank. The repair work was performed at a height of 240 cm from ground. As the resin hopper tank is located relatively higher than the other components, its external dose rate is dominated by the effect of the 1000 kg resin distributed in the resin hopper tank, indicating a high dose rate around this tank.

3.1.1. Repairing spent resin mixture separator

The external dose rate received by the workers while repairing the spent resin mixture separator was 1.64E-01 mSv/h. ^{137}Cs, included in the spent resin mixture separator, has the greatest impact as shown in Table 2 (about 72% of total). The maximum workable time, derived within the worker's dose limit without considering internal exposure, was confirmed to be 125 h, and thus, repairs can be performed for 15.6 days in a year as shown in Table 3.

3.2. Repairing zeolite and activated carbon storage tank

The external dose rate received by the workers while repairing the zeolite and activated carbon storage tank was 7.23E-01 mSv/h. ^{137}Cs, included in the zeolite and activated carbon storage tank, has the greatest impact as shown in Table 2 (about 94% of total). The maximum workable time, derived within the worker's dose limit without considering internal exposure, was confirmed to be 27.7 h, and thus, repairs can be performed for 3.5 days in a year as shown in Table 3.

3.3. Repairing resin hopper tank

The external dose rate received by the workers while repairing the resin hopper tank was 5.78E-01 mSv/h. ^{137}Cs, included in the resin hopper tank, has the greatest impact as shown in Table 2 (about 63% of total). The maximum workable time, derived within the worker's dose limit without considering internal exposure, was confirmed to be 38.5 h, and thus, repairs can be performed for 4.8 days in a year as shown in Table 3.

Table 2
Dose rate and dose rate % for each component failure.

Source name (Spent resin mixture separator)	Dose rate (mSv/h)	Dose rate %
^{137}Cs	1.19E-01	72.26
^{60}Co	3.09E-02	18.83
^{152}Eu	1.11E-02	6.77
Etc. (^{57}Co,^{51}Cr,^{134}Cs,^{54}Mn,^{95}Nb,^{125}Sb,^{95}Zr,^{154}Eu)	3.50E-03	2.13
Total	1.64E-01	100.00
Source name (Zeolite and activated carbon storage tank)	**Dose rate (mSv/h)**	**Dose rate %**
^{137}Cs	6.80E-01	94.04
^{60}Co	3.80E-02	5.25
^{152}Eu	2.08E-03	0.29
Etc. (^{57}Co,^{51}Cr,^{134}Cs,^{54}Mn,^{95}Nb,^{125}Sb,^{95}Zr,^{154}Eu)	3.04E-03	0.42
Total	7.23E-01	100.00
Source name (Resin hopper tank)	**Dose rate (mSv/h)**	**Dose rate %**
^{137}Cs	3.64E-01	63.09
^{60}Co	1.42E-01	24.61
^{152}Eu	5.31E-02	9.20
Etc. (^{57}Co,^{51}Cr,^{134}Cs,^{54}Mn,^{95}Nb,^{125}Sb,^{95}Zr,^{154}Eu)	1.79E-02	3.11
Total	5.78E-01	100.00
Source name (Microwave device)	**Dose rate (mSv/h)**	**Dose rate %**
^{137}Cs	4.17E-02	52.82
^{60}Co	2.60E-02	32.95
^{152}Eu	8.86E-03	11.23
Etc. (^{57}Co,^{51}Cr,^{134}Cs,^{54}Mn,^{95}Nb,^{125}Sb,^{95}Zr,^{154}Eu)	2.36E-03	2.99
Total	7.89E-02	100.00
Source name (^{14}C detached resin storage tank)	**Dose rate (mSv/h)**	**Dose rate %**
^{137}Cs	3.57E-02	48.06
^{60}Co	2.71E-02	36.51
^{152}Eu	9.13E-03	12.30
Etc. (^{57}Co,^{51}Cr,^{134}Cs,^{54}Mn,^{95}Nb,^{125}Sb,^{95}Zr,^{154}Eu)	2.33E-03	3.13
Total	7.42E-02	100.00

Table 3
Maximum workable time and days for each device.

	Spent resin mixture separator	Zeolite and activated carbon storage tank	Resin hopper tank	Microwave device	^{14}C detached resin storage tank
Maximum workable time (h)	125	27.7	38.5	250	263.2
Maximum workable days (d, 8 h a day)	15.6	3.5	4.8	31.2	32.9

3.4. Repairing microwave device

The external dose rate received by the workers while repairing the microwave device was 7.89E-02 mSv/h. ^{137}Cs, included in the microwave device, has the greatest impact as shown in Table 2 (about 53% of total). The maximum workable time, derived within the worker's dose limit without considering internal exposure, was confirmed to be 250 h, and thus, repairs can be performed for 31.2 days in a year as shown in Table 3.

3.5. Repairing ^{14}C-detached resin storage tank

The external dose rate received by the workers while repairing the ^{14}C-detached resin storage tank was 7.42E-02 mSv/h. ^{137}Cs, included in the microwave device, has the greatest impact as shown in Table 2 (about 48% of total). The maximum workable time, derived within the worker's dose limit without considering internal exposure, was confirmed to be 263.2 h, and thus, repairs can be performed for 32.9 days in a year as shown in Table 3.

3.6. Assessment of internal dose from spent resin treatment device during repair

When considering the repairing of the spent resin mixture separator, it was assumed that a worker wearing an APF 50 air-purifying respirator inhales 2% of the nuclides during the repair work. The largest and smallest internal doses received by the workers were 2.31 E+03 and 2.72 E+02 mSv from the zeolite and activated carbon storage tank and the spent resin mixture separator, respectively. These values are very large compared to the annual dose limit of 20 mSv for workers. To ensure that workers do not receive an internal dose, it is therefore necessary to design a tank that can be repaired externally in case of device failure or to store the internal spent resin mixture separately. For example, a direct contact between the worker and the resin mixture can be prevented by constructing pipes and pumps through which the spent resin mixture, inside the treatment device, can be transported back to the existing spent resin mixture storage tank during the repair work period. This will significantly reduce the internal exposure of the worker since it does not fundamentally cause conditions for inhaling radionuclides contaminated in the resin.

3.7. Radiological safety of spent resin treatment device during repair

Although internal dose results were derived conservatively, very large internal dose values ranging from a maximum of 2.31 E+03 mSv to a minimum of 2.72 E+02 mSv were obtained. To reduce this value, extra pumps and tanks are needed to temporarily store the spent resin mixture in the failed component. To ensure the maximum repair time, it is best to reduce the internal dose to 0 mSv. That is, if it is necessary to check the inside of components during the repair work, then the entire spent resin mixture contained in the device should be moved to another place. The most efficient design is to enable repairs outside each device. Table 3 shows the maximum possible repair time and repair days in a year obtained for repairs performed outside the device.

4. Conclusion

Radiological safety evaluations were performed for workers performing repairs on the components of a spent resin treatment device with a spent resin mixture treatment capacity of 1 ton per day. When each component was designed to be repairable from the outside, the worker received only an external dose for a maximum of 32.9 days (^{14}C-detached resin storage tank) and a minimum of 3.5 days (zeolite and activated carbon storage tank) in one year. The average number of maximum repair days for the five components is 17.6 days (4.82%) in one year. However, if it is necessary to check the inside of a malfunctioning device, then the worker receives a very large internal dose (2.72 E+02 to 2.31 E+03 mSv). To ensure radiological safety while workers repair the device, extra pumps and tanks are necessary to temporarily store the untreated spent resin mixture contained in the devices before starting the repairs.

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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