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[blurb]A Solid low level radioactive waste (LLRW) from radioisotope research usage is to have an uneven distribution of RI. The RI quantity in Solid LLRW was determined by an experiment incineration of Solid LLRW. We examined whether it is possible to Clearance of Solid LLRW as compared to international standards.

[t]Evaluation of IAEA Clearance Concept for Low-level Radioactive Waste from a Radioisotope Research Institute

[a]Yasuhiro Yumoto,* Shigeru Okada,[dagger] Ikuo Kinno,* Tomohiro Nagamatsu,* Kazuhiro Nouso,[ddagger] and Eiichi Nakayama*

[abs][fnc,1]The clearance of solid low-level radioactive laboratory waste (LLRW) after decay-in-storage (DIS) obtained from a research institute and thoroughly separated using the separation and classification protocols presented in this study was evaluated. Method: The radioisotope (RI)

content of incinerated LLRW from the specified RI research group (group A); the RI content of LLRW obtained in fiscal year 2000, which contained radionuclides with half-lives of less than 164 d (LLRW2); and the RI content of the LLRW reported in group A's disposal records were compared. The LLRW2 and LLRW of group A were incinerated after 2 y of decay-in-storage and immediately after storage, respectively. Results: The highest ratio of the RI of incinerated LLRW to the value in the disposal records was 2.52 for ^{51}Cr. The radioactivities of radionuclides in both the LLRW2 and LLRW for ^{35}S, ^{45}Ca, ^{51}Cr, ^{125}I, ^{32}P, ^{33}P, and ^{99m}Tc and the incinerated ash after 1 y later of decay-in-storage were below the clearance level defined by the RS-G[hyphen] 1.7 of the International Basic Safety Standard without contamination by ^{3}H and ^{14}C. These remains contained very small amounts of some long-half-life radionuclides of natural origin after 7 y of decay-in-storage. Conclusion: This LLRW separation protocol was effective for the separation of ^{3}H and ^{14}C. LLRW2 after 2 years of DIS and its incinerated ash after one year later of DIS were below the clearance level for radioactivity and radioactivity concentration.

[key] operational topics; waste, low-level; waste disposal; waste management

[intro]

[txt]The management of low-level radioactive waste (LLRW) generated by biochemical research and diagnostic patient-care procedures typically involves a series of steps. Most of these materials contain short-lived radioactive materials and are allowed to decay in storage (DIS) to essentially nonradioactive materials before disposal (NRC 2001). The institute can easily store the short-lived radionuclides to reduce the radioactivity level below the IAEA clearance values. This practice is performed around the world.

[p]Recently, the safety regulations for LLRW have been reviewed by the Japan Ray Council (mid [hyphen] 2006), which ultimately agreed with the international clearance level. Act to amend the

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[A] Add introductory sentence to follow title.

law on the prevention of radiation hazards due to radioisotopes was promulgated on 10 May 2010. Enforcement of the law will be carried out after a technical study. In Japan, Ordinance No. 30 of the Ministry of Health and Welfare establishes that solid waste containing radionuclides having an ultra-short half-life used in positron emission tomography (PET) may be removed from controlled areas after 7 d of decay-in-storage in disposal facilities; however, this regulation is not applied to RI waste in research institutes.

[p]Some combustible and semi-combustible LLRW in research institutes have been collected and incinerated by the Japan Isotope Society Corporation (JRIA), and the ash has been stored (JNSC 2004; JRIA 1992[B], 2006). LLRWs were separated and segregated by a new protocol that was modified from the LLRW classification table (JRIA 2006). The specified RI experiments were performed using a single nuclide as the tracer. The consistency of the amount of RI recorded as LLRW by the workers who conducted the specified RI experiments, which were performed using a single nuclide as the tracer, and that by employees of the RIC in fiscal year 2000 were evaluated by identifying and quantifying the RI obtained after incineration. Moreover, the radioactivity and radioactive concentration levels of incineration ash were compared to the clearance level of RS-G[hyphen] 1.7 (IAEA 2004). The dioxin level after incineration was below the clearance levels in our study, but the RI waste may contain harmful chemicals.

[h1]MATERIALS AND METHODS

[txt]Workers classified LLRW according to the separation and classification protocol in experiment room of laboratories, placed it in the corresponding plastic bag for each RI waste during examination, and disposed of it. The LLRW was stored in a radioactive waste storage facility. All workers were requested to record the amount of radionuclides used and the estimated amounts in

[B] Add complete citation for JRIA 1992 to ref list.

the waste using the radiation control computer. Table [tbc] 1 shows the annual total amounts of RI purchased and the amount that carried over from the previous year between 1996 and 2000 at the RIC. The radioactivities of RI in each radionuclide in the collected LLRW are the sum of the radioactivities in the waste disposal records reported by the workers.

[h2]LLRW classification and storage methods used by group A

[txt]LRW containing ^{32}P, ^{35}S, ^{45}Ca, ^{51}Cr, ^{123}I, ^{125}I, and ^{99m}Tc were stored separately in group-A drums AP, AS, ACa, ACr, AI, and ATc, respectively.

[p]LLRW containing ^{3}H and/or ^{14}C was stored separately in group-B drums BH and BC, respectively.

[p]In fiscal year 2000, LLRW containing radionuclides with half-lives of more than 164 d was stored by unspecified RI workers in drum C and drum E, separately from other radionuclides with half-lives of less than 164 d (LLRW2). The LLRW2 drums included drum D, containing only ^{45}Ca (Table [tbc] 2).

[p]The group A LLRWs were stored after collection and incinerated immediately after storage. LLRW2 was incinerated after 2 years of decay-in-storage. The type and the radioactivity of the RI of LLRW stored by the specified RI workers and those of LLRW2 were compared with those of the incinerated LLRW and those of LLRW2 (Tables 2 and [tbc] 3).

[h2]Method used in the incineration experiments

[txt]In a typical incineration, LLRWs are placed in a high-temperature (over 900°C) environment and burned to ash. The concentration of gaseous RI in the smoke dust and at the final exhaust port

was measured during incineration. The RI in the gas and the RI absorbed by small particles were sampled from the smoke duct by suction at the same flow rate as that of the exhaust in the smoke duct using a stainless-steel tube with a diameter of 2 cm fixed perpendicular to the smoke duct. After trapping the RI with a CP20 activated carbon filter and a GB-100R filter (Toyoroshi Co. Ltd., Tokyo, Japan), the RI in the uptake gas was liquid trapped by 5% $Na_2S_2O_3$, 0.5 M HCl, 0.5 M NaOH, 0.5 M H_2O_2, and Carbosorb (Perkin Elmer Inc. Co. Ltd., MA), and the cold traps were examined using a gamma-ray spectrometer (Auto-well GAMMA system ARC[hyphen]2000; Aloka Co. Ltd, Tokyo, Japan), a pure germanium semi-conductor detector (E 6G; Seiko Co. Ltd, Tokyo, Japan), and a liquid scintillation counter (TRI-CARB 2300TR; Packard Co. Ltd, Tokyo, Japan) (Yumoto et al. 2000).

[p]The radioactivity and radioactivity concentration of furnace ash, ash on the incinerator wall and ash in the cyclone after incineration of LLRW containing ^{32}P, ^{35}S, ^{45}Ca, ^{51}Cr, ^{123}I, ^{125}I, and ^{99m}Tc in group A and LLRW2, including drum D, which only contained ^{45}Ca, were measured after incinerating the samples that were immediately obtained and those that had been stored for 7 y (Tables 2 and 3).

[p]The radioactivity of the furnace ash, the ash on the incinerator wall and the ash in the cyclone were examined using a gamma-ray spectrometer, a germanium semi-conductor detector, a low-energy beta-ray detector and a bio-imaging analyzer (BAS [hyphen] 2000II; Fuji Photo Film Co., Ltd, 2-26-30, Nishiazabu, Minato-ku, Tokyo 106-8620, Japan), which showed a 0.53 Bq g [minus] ^{1} detectability for ^{14}C. ^3H and ^{14}C in the furnace ash, furnace-wall ash, and cyclone ash were measured with an automatic sample combustion system (ASC [hyphen] 113; Aloka Co. Ltd. Tokyo, Japan) using Carbosord, a cold trap and a liquid scintillation counter (Yumoto et al. 1999, 2000). The detectabilities of these apparatus with respect to ^3H and ^{14}C are 0.10 Bq g [minus] ^{1} and 0.05 Bq g

[minus] 1, respectively. The recovery of ^{3}H and ^{14}C was 96.3% and 90.6%, respectively (Yumoto et al. 2000).

[p]A HEPA filter for high-temperature applications (ASTE-Z[hyphen] 60E; Nihonmuki Co. Ltd, Tokyo, Japan) was fixed at the final exhaust port. The RI concentrations in the final exhaust port after HEPA filtration were measured by a gamma-ray monitor (DGM[hyphen]101; Hitachi Aloka Medical, Co., Ltd., 6-22-1, Murei, Mitaka City, Tokyo, Japan), an iodine monitor (DDM[hyphen]201; Aloka Co. Ltd) and a dust monitor (DDM[hyphen]151; Aloka Co. Ltd) in our RIC (Fig. [fgc] 1).

[h1]RESULTS AND DISCUSSION

[h2]Evaluation of the disposal method of LLRW

[txt]The accuracy of the LLRW data reported by workers after tracer experiments was evaluated by comparison with the data obtained from the incineration experiments. Because ^{3}H, ^{14}C, ^{35}S and radio-iodine (^{125}I, ^{131}I) are volatile, these radionuclides cannot be removed by a duct collector. The radioactivity of the gas in the smoke duct passing through the cyclone was measured during incineration. These radioactivities were below the legal limit, and the radioactivity concentration in the smoke duct after HEPA filtration was also below the legal limit at the final exhaust port in all incineration experiments (Japan Law 1998).

[p]LLRW incineration experiments conducted by RI research group A classified using this method showed that the waste contained the used radionuclide but did not contain ^{3}H and ^{14}C (Table 3).

[p]The radioactivity concentration of ^{14}C in the LLRW of RI group B was 12.3±7.5 Bq g [minus] 1 (*n* = 11), which is greater than that of BS-G[hyphen] 1.7. Upon incineration at 900-1,100°C, the

radioactivity concentration of ^{14}C showed the background level in the furnace ash and wall ash and 1.82±0.92 Bq g[minus]1 in the cyclone ash (n = 11) (Fig. [fgc]2). These radioactive ashes were stored in 2 steel containers. Nogawa reported that the radioactivity of ^{3}H and ^{14}C was not observed in the liquid scintillation waste residue (Nogawa et al. 1997) and that no radioactivity in the incineration residue was confirmed, even for radioactive organic waste fluid (Emery et al. 1992). Incombustible aluminum, stainless clips, and glass were mixed with the combustible waste. In the furnace ash, furnace wall ash and cyclone ash after incineration of LLRW2, ^{3}H or ^{14}C could not be detected using an automatic sample combustion system with Carbosorb and a cold trap (ASC [hyphen] 113; Aloka Co. Ltd.) with a scintillation counter or imaging analyzer after 7 y of decay-in-storage. The incineration experiments demonstrated that our new separation protocol for LLRW was effective in separating ^{3}H and ^{14}C. Neither ^{3}H nor ^{14}C were significantly mixed in the LLRW2 containing very small amounts of a long-half-life radionuclide of natural origin below the RS-G[hyphen] 1.7 levels (Table 3), which was not used in our laboratory.

[p]The guarantee of environmental safety is invaluable at clearance time. A very small amount of 8 long-half-life non-artificial radionuclides (less than BS-G[hyphen] 1.7) was detected by the germanium semi-conductor detector, automatic sample combustion system, and liquid scintillation counter in the furnace ash, ash on the incinerator wall, and cyclone ash. These radionuclide levels were below the clearance values of RS-G[hyphen] 1.7 (Table 3) (IAEA 1996, 2004).

[p]The $BR^{[minus]\,1}$ ratio, which is the ratio of the amount of RI (B) in group A determined by experimental incineration to that reported in the disposal records (R) by group-A workers, was high (1.36-2.52) for ^{3}H, ^{35}S, ^{45}Ca, ^{51}Cr, and ^{125}I, indicating that the incineration experiments yielded higher values than had been recorded at disposal, and low (0.41-0.45) for ^{14}C, ^{32}P and ^{33}P. The highest $BR^{[minus]\,1}$ ratio obtained by the separation and classification method was 2.52 in the ACr

group, showing 1.78±0.59 (*n* = 12). The value was corrected by the 87.7% recovery rate (Yumoto et al. 2000).

[h2]Study of the practicality of LLRW clearance that was accurately separated and classified using the new method

[txt]The rate of weight reduction by incineration at 900-1,050ºC was more than 98.1%. The incineration ash, ash on the incinerator wall, and cyclone ash of the LLRW in the AP, ACr, ACa, AI, ATc, and AS groups were stored for 1 y more to decay. The RI level of these ash samples was below the BS-G[hyphen] 1.7 value (Table 3).

[p]The radioactivity concentrations of LLRW2 in fiscal year 2000 were below the BS-G[hyphen] 1.7 value after 2 y of storage. In contrast, the radioactivity concentration of ^{51}Cr, ^{125}I and ^{35}S in the incineration ash, which was greater than the BS-G[hyphen] 1.7 values after 2 y of storage, was less than the BS-G[hyphen] 1.7 value after 3 year of storage (Table 2).[C] The clearance of the incineration ash of LLRW2 may be possible by storing the ash for 3 y, after which no significant contamination by radionuclides with half-lives longer than the clearance level was confirmed by incineration experiments performed at the Radioisotope Research Institute.

[h2]Calculated decay time when the annual stock quantity in an RI storehouse reaches clearance level

[txt]The total radioactivity concentrations of radionuclides with half-lives of less than 90 d in the annual stock from 1998 to 2000 were lower than the clearance level after 3 y of decay-in-storage. In addition, the total radioactivity of radionuclides with half-lives of less than 60 d and radioactivity concentration in the RIC storeroom after 2 y of decay-in-storage were below the clearance level

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[C] Please verify: In contrast, the radioactivity concentration of ^{51}Cr, ^{125}I and ^{35}S in the incineration ash, which was greater than the BS-G[hyphen]1.7 values after 2 y of storage, was less than the BS-G[hyphen]1.7 value after one more year of storage (Table 2).

(Table 1). The radioactivity concentration of 8 nuclides in group A was lower than the BS-G[hyphen] 1.7 levels after 1 y of decay-in-storage (Table 2). In contrast, the Japan Radioisotope Association (JRIA), which collected LLRW with a short half-life, reported that ^{32}P with less than a 30[hyphen]d half-life after one year of decay-in-storage, ^{125}I with less than a 60[hyphen]d half-life after 2 y of decay-in-storage, and ^{35}S with less than a 90[hyphen]d half-life after 3 y of decay-in-storage showed annual radioactivity values below the BS-G[hyphen]1.7 level (JRIA 2006). Because the amount of ^{45}Ca found in the LLRW studied at our institute was small, three years of decay-in-storage of nuclides with a half-life of 164 d was performed. If the radioactivity concentration of ^{45}Ca in LLRW is greater than 356 Bq g $^{[hyphen]\ 1}$, nuclides in the LLRW with a 90[hyphen] d half-life will be stored for 3 y.

[p]A guarantee of environmental safety is indispensable at clearance time. A very small amount of 8 long half-life non-artificial radionuclides (less than RS-G[hyphen] 1.7) was detected by a germanium semi-conductor detector, an automatic sample combustion system and a liquid scintillation counter in the furnace ash, ash on the incinerator wall, and cyclone ash by incineration of LLRW2 after decay-in-storage for 7 y. These radionuclide levels were below the clearance values of RS-G[hyphen] 1.7 (Table 3).

[p]In Europe and United States, radioactive waste at radioactivity levels below a certain level is not controlled and can be treated as general waste (EC 2001; Krieger et al. 2002). According to United States regulations (10CFR Part 35.92, U.S. NRC 2003), solid radionuclide waste with a half-life shorter than 120 d used in the medical field is stored for a sufficiently long period and can be disposed of after background-level radioactivity is confirmed using an appropriate measuring apparatus. In Japan, solid waste containing radionuclides with ultra-short half-lives used in PET may be removed from controlled areas after 7 d in storage and disposal facilities; however, this

regulation does not apply to solid LLRW generated by research institutes. The RI level of solid LLRW2 that did not contain ^{3}H and ^{14}C in fiscal year 2000 sorted by our separation and classification method decayed in 2 y of storage to the clearance values, which are below the clearance level of the IAEA (2004, 1996). Table 3 indicates that the waste segregation of ^{3}H and/or ^{14}C was successful: there were no long-lived radionuclides ($^{3}H/^{14}C$) in the short-lived radionuclide waste. In addition, Table 3 indicates that the incineration of $^{3}H/^{14}C$ waste does not lead to concentrations of long-lived naturally occurring radionuclides above the IAEA clearance values. Therefore, it is possible to incinerate $^{3}H/^{14}C$ waste to dispose of the radionuclides to the atmosphere and reduce the volume of material that must be handled as radioactive waste. If amounts of longer-half-life radionuclides above the clearance level are mixed in LLRW, this LLRW will be deposited in shallow land. In Germany, the clearance values of RI wastes at research institutes are regarded as the permissible levels.

[h1]CONCLUSION

[txt]Radioisotope workers classified solid LLRW according to the separation and classification protocol used in the laboratory.

[p]LLRW2 containing radionuclides with half-lives of less than 164 d were stored separately from other radionuclides. This LLRW separation protocol was effective for the separation of ^{3}H and ^{14}C. After 2 y and 1 y of decay-in-storage, respectively, LLRW2 and its incinerated ash showed radioactivity concentrations below the clearance level.

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experiment and for useful comments. We also thank Nakamura Takashi, RIC of Tohoku University, for his useful comments.

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[E] Clarify "Law to amend some of the legislation on the prevention of radiation hazards due to radioisotopes. May 10, 2010." Give complete citation and call out in text using (Author Year).

[F] Add city of publication to Mobbs and Harvey 1999.

[G] Clarify Middle Report 2006. Cite in text using (Author Year). Clarify citation: Author. Title. City of publication: publisher; year of publication.

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[fnote]

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[fig2]1 Combustion furnace equipped with two burners powered by city gas or propane gas, a measurement system for radioactive exhaust gas, and a sampling filter for smoke duct gas and cyclone separator ash. The furnace bed space is 0.49 m^2 with fire-brick tile walls. The incineration temperature of the combustible and semi-combustible LLRW reached 900°C to 1050°C. A heat-resistant high-efficiency particulate activity (HEPA) filter was set on top of the smoke duct, which was equipped with an air suction fan. The terminal smoke duct gas was aspirated from the terminal smoke duct; cooled to below 35°C with flowing water, and measured with a detector. The sampling gas was sucked from the smoke duct after the cyclone separator and passed through the cooling unit and a sampling filter in the automatic sample changing apparatus for smoke duct gas. The smoke duct gas that passed through the sampling filter was passed through the liquid-collection apparatus containing 5% $Na_2S_2O_3$, 0.5 N HCl, 0.5 N NaOH, 0.5 M H_2O_2, Carbosorb, and a cold trap for radionuclides corresponding to each nuclide. The liquid sample was measured with a liquid scintillation counter or a gamma scintillation spectrometer.

[fig1]2 Evaluation of ^{14}C radioactivity in the furnace ash, wall ash and cyclone ash after 7 y of Decay-in-storage.

Ash samples from the furnace, furnace wall, and cyclone ash were absorbed on GB100RTM filters by a dust sampler. Ash samples in which the sample thickness did not exceed 4mg cm

[minus] ^{2} were used for image analysis. The image analysis shows the ^{14}C radioactivity of the cyclone ash (1), furnace ash (2), and wall ash (3) which were incineration ash of LLRW in the dram can BC (Table 3.) without containing of ^{40}K using a pure germanium semiconductor detector. ^{14}C level in these ash samples (1), (2) and (3) showed BG level. And also, ^{40}K activities in these incineration ash samples (1), (2) and (3) showed BG level by a pure germanium semi-conductor detector. The cyclone ash (4), which was obtained from the combustion of LLRW from the specified Group BC using ^{14}C, contained 732 mg of ash, and showed 2.0 Bq of ^{14}C. A ^{14}C radioactivity concentration of 2.73Bq $g^{-1}$was detected after one year of decay-in-storage.
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