Characterization and Classification of Radioactive Wastes from Disposal Silo

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Abstract
In the present work, classification of radioactive wastes based on Annual Intake (AI) values is studied. Where the characterization of radionuclides was done by hand held GeLi detector with an overall efficiency better than 42%. It was noted the most predominant contaminant are Cs-137, Co-60 and Pa-234.The radioactive waste in disposal silo has been divided into five categories according to the harmful effect of radionuclides.For the purpose of storageradioactive wastein a safe manner, it wassuggesteda new method by shielding radioactive waste in each category with concrete;where the thickness of shielding is the time required to reduce the annual dose to 10%.

Key words: Annual Intake, classification of radioactive wastes, disposal silo

توصيف وتصنيف النفايات المشعة في مخازن الطمر

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الخلاصة:
فى هذا البحث، تم تصنيف النفايات المشعة المخزونة بالقرب من السطح على أساس قيم التناول السنوي (AI).وتحقيقا لذلك تم توصيف النويدات المشعة باستعمال كاشف الجرمانيوم–ليثيوموذو كفاءة كلية تزيدعلى42٪. وقد لوحظ أن معظم الملوثات هي النويدات المشعة Cs-137 و Co-60 و Pa-234. تم تصنيف النفايات المشعة الموجودة في مخزن الطمر الى خمس فئات وحسب قيم التناول السنوي.ولغرض المعالجة اقترحت طريقة جديدة نضمن من خلالها خفضمستوى الاشعاع الى مستويات غير ضارة بواسطة تدريع النفايات المشعة في كل فئة بخرسانة؛ حيث ان سمك التدريع هو الوقت اللازم لخفضالجرعة السنوية إلى 10٪ من قيمتها الابتدائية.

Introduction

Radioactive waste classification systems have been developed to enable wastes having similar hazards to be grouped for purposes of storage, treatment, packaging, transportation, and/or disposal. As recommended in the National Council on Radiation Protection and Measurements' Report No. 139[1].However, the level of waste is not the only indicator of radionuclide dangerous. The activity, energy and the type of the ionizing radiation emitted by a pure radioactive substance are important factors in determine how dangerous it will be. In addition, the chemical properties of the radioactive element will determine how mobile the content is and how probably it is to spread into the environment and affect human bodies. This is further complicated by the fact that many radioisotopes

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do not decay immediately to a stable state but rather to a radioactive decay product leading to decay chains[2].

Risk based classification of radioactive and risk chemical wastes, a preferred classification system would be based mainly on the health risks to the public that arise from waste disposal and secondarily on other properties such as the short term processes of managing a waste, i.e., the waste classification system would be risk informed. Schemes for the management of radioactive wastes are varied according to their classification. Nevertheless, classification may be developed from different rules, such as; activity, half-life, physical and chemical form, operational or long term safety, therequirements of process engineering, the availability of management or disposal facilities or the source of waste generation[3]. Different systems have developed in order to classify radioactive wastes according to the national policies and strategy plans that are related to particular country in which radioactive waste is managed Regulatory or technical operators, should base the classification systems on the following factors: 1) the radioactive waste component, and its concentration; 2) limits and conditions set by the authorities; 3) Disposal system and design; 4) pathways or scenarios prescribed for safety assessments; 5) operational restrictions; 6) site specific conditions; 7) social or political aspects; and 8) legal definitions and requirements [4,5].

These factors may restrict the degree of freedom for the selection and put of a classification system. Accordingly, all these factors have to be evaluated before the classification system can be derived in spite of these factors are changed case by case. ALI is the is defined as the smaller value of intake of a given radionuclide in a year by the reference man that would result in a committed effective dose calculated by the International Commission on Radiological Protection (ICRP). On the other hand, radiotoxicity is a measure the extent of harmful a radionuclide is to human health when inhaled or ingested. Radiotoxicity depends on the type and energy of the radiation emitted and the radionuclide's biochemical behavior in the human body. The harm that can be done depends on the dose of radiation received. The paper illustrates a new proposal or approach to classify the waste for near surface disposal. The new approach is based on to what extent the radionuclide element hazard for human according to its ALI value. Boundary levels between classes are displayed as orders of magnitude and typical characteristics of waste classes. The classification systems are as follow;

1. Exempt waste (EW):Waste that meets the criteria for exemption from regulatorycontrol for radiation protection purposes.
2. Very short lived waste (VSLW): and
3. Very low level waste (VLLW**):**
4. Low level waste (LLW):
5. Intermediate level waste (ILW):
6. High level waste (HLW) [6].

Treatment of radioactive waste may involve segregation, chemical adjustment, decontamination, containment, volume reduction, removal of radio nuclides from the waste, and change of composition. Conditioning may involve conversion of the waste into a solid form, cementation, verification, enclosure of waste in containers, and provision of an over-pack for transport. Operational management involves categorization of the waste based on its physical and chemical characteristics.

The categories are loosely related to occupational risk. Classification of radioactive waste for disposal is conversely related to long-term risks to members of the public and the environment [7,8].

Safety Case and Safety Assessment

The primary purposes of the safety assessment are to determine whether an adequate level of safety has been achieved for a facility or activity and whether the basic safety objectives and safety criteria established by the designer, the operator and the regulatory body have been fulfilled. For a disposal facility, safety assessment entails evaluating the performance of the disposal system and quantifying its potential radiological impact on human health and the environment. Safety assessment is one component of the safety case for a disposal facility and should consider the possible radiological impacts of the facility both during its operation and in the post-closure phase. Radiological impacts may arise from gradual processes which may cause the facility and its components (e.g. barriers) to degrade, and from discrete events that may affect the isolation of the waste (e.g. earthquakes, tsunamis, floods, fire, inadvertent human intrusion).

Dose Conversion Factors (h) provided by the International Atomic Energy Agency (IAEA) in its Safety Series 115, are available from this site for viewing or download. These factors give the dose per unit intake by inhalation or ingestion for a large number of radionuclides, for adults and children of various ages. They are thus useful for calculating committed doses for workers, who might experience intakes in the workplace, as well as individuals or populations near nuclear sites, where intakes might occur due to offsite releases of radionuclides.

According to the IAEA Safety guide [3], depending on the purpose of a radioactive waste classification system there exist different approaches to its derivation. One basic method of classification is a qualitative description of the individual classes. In this case, mostly general characteristics of the radioactive waste are used as criteria for the classification. Nonetheless, numerical values to characterize broad band's or orders of magnitude may also be helpful for classification by this approach. The other method is a quantitative approach, i.e. numerical values are given for the definition of most classes. Additionally, the safety requirements publication, Predisposal Management of Radioactive Waste [5], requires that: “At various steps in the predisposal management of radioactive waste, the radioactive waste shall be characterized and classified in accordance with requirements established or approved by the regulatory body” (Requirement 9). This is to ensure that proper and adequate provision is made for the safety implications associated with the management and disposal of the waste. The purpose of this research is to classification and treatment the near-surface disposal radioactive waste in a way which ensures that there is no unacceptable risk to humans.

Experimental details

The Annual Intake, *AI*, is the intake resulting in a committed effective dose that is equal to the annual dose. The unit for the Annual Intake is the Becquerel (Bq), and is calculated as follows:

$$AI=\frac{D_{AL}}{h} \qquad (1)$$

where$D_{AL}$is the annual dose limit for the effective dose, and$h$ is the dose conversion factor. The equivalent dose $H_{T,R}$ in tissue or organ T is obtained by multiplying the average absorbed dose $D_{T,R}$in the tissue or organ by a radiation weighting factor W_R*[9]*:

$$H_{T,R} = D_{T,R} \times W_{T,R} \qquad (2)$$

where$W_R$is the radiation weighting factor for radiation quality R,

If the radiation is composed of several radiation qualities with different $W_R$values, the equivalent dose $H_T$is:

$$H_T = \sum_R D_{T,R} \times W_R \qquad (3)$$

The unit of equivalent dose is the Sievert (Sv).

The effective dose $D_{AL}$is the sum of the equivalent doses H_T, multiplied by the tissue weighting factors W_T:

$$D_{AL} = \sum_T H_T \times W_T \qquad (4)$$

$$D_{AL} = \sum_T W_T \sum_R D_{T,R} \times W_R \qquad (5)$$

The equivalent dose is used to estimate the harmful effects of radiation in a certain tissue [10]. The effective dose is chiefly used to estimate the risk of stochastic harmful effects of radiation on an individual and its values can be calculated by measuring the activity of each radionuclides in the waste.

Gamma spectrometer and relevant accessories were supplied by Canberra, USA used to measure the activity concentrations for each radionuclidesin the silo.

The silo was re-equipped with 5 ton bridge crane to enable the removal of the concert plugs of the wells and cover the RW. The wells were numbered and a dose map for the top and the sides of the facility were drawn. The radiation dose rate for each well were measured before and after the removal of the concrete plugs, each well were identified for the radionuclide that is contaminating the waste, ithas been used a very sophisticated, well advanced piece of equipment produced by ORTEC so called (hand held GeLi detector) with an overall efficiency (better than 42%). More than 95% of the concrete wells were a converted. The resolution of this detector is 1.32185 MeV for Co-60 energy.The energy calibration of Ge(Li) gamma-ray spectrometer is performed by Co-60 radioactive source as shown in figure-1 .

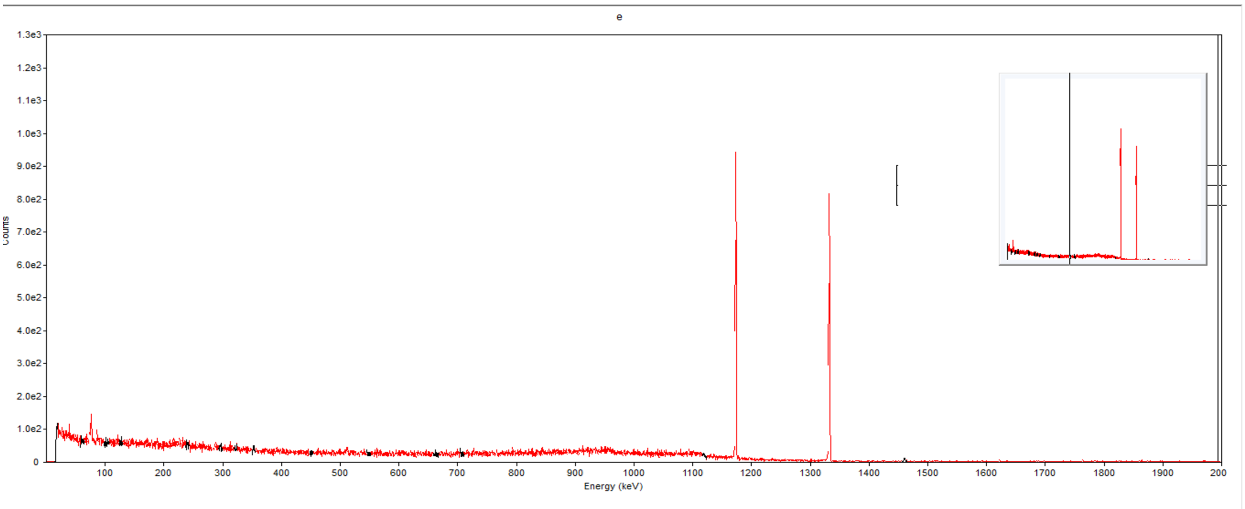

Figure. 1- Calibration spectrum for GeLi detector using Co-60

The samples spectra are corrected for background radiation, the background values obtained by using Eq.6:

$$\text{Background (Bq)} = \frac{Area}{IY\%\ EFF\ \%\ Tc} \qquad (6)$$

where
Area: The neat area under the peakcount
I_Y %: The branching ratio for photon energy
Eff: Efficiency of the Ge(Li) detector.
Tc:The total counting time interval in seconds

The specific activity for samples has been measured using:

$$\text{Specific Activity(Bq)} = \frac{(Area-Bg)/Tc}{IY\%\ Eff\ \%\ m} \qquad (7)$$

where m=The mass of samples

Results and Discussions

Table-1 shows the gate numbers, types, energy, half-lives and dose of radionuclides for public. The annual effective dose are obtained by using Eq.(5). Finally the AI values are obtained by using Eq. (1).

Table 1- Types of radionuclides, the thickness of concrete shielding (x) and the time (t) required to reduce the effective dose to 10%

No.	Type of radionuclide's	Energy keV	Half life	μ/ρ (cm^2/g)	X cm	Annual effective of radionuclide μSv	H[10] Sv/Bq	AI Bq	t
1	Co-60	1173.6	5.2714 y	0.0587	17.787	2.018	5.20E-09	388	17.50 y
2	Cs-137 Eu-152 Pa-234M Co- 60 Y-88 Bi-214	661 778.90 1001.27 1173 1382.20 1764.49	30.07 y 13.537 y 6.70 h 5.2714 y 107d 0.332 h	0.0762 0.0709 0.0636 0.0587 0.0552 0.0496	13.714 14.739 16.431 17.803 18.931 21.069	23.532 8.508 210.07 113.172 162.75 1.23	4.60E-09 4.20E-08 3.80E-10 5.20E-09 4.10E-09 7.10E-09	5115 202 552829 21763 39695 173	100 y 44.93y 22.24h 17.5 y 355.13d 1.10h
3	Pa-234M U-235	98.5 185.8	6.70 h 7.04E+0	0.175 0.133	5.971 7.857	776.12 0.75	3.80E-10	2042415 1.44	22.24h 23.37E+0

	Eu-152 Sb-122 Cs-137 Co -60	344.32 615 661 1173	8 y 13.537 y 2.70 d 30.07 y 5.2714 y	0.101 0.0786 0.0762 0.0587	10.347 13.295 13.714 17.802	6.487 7216.8 15.816 25.47	5.20E-07 4.20E-08 3.60E-10 4.60E-09 5.20E-09	154 20046666 3438 4898	8 y 44.93y 8.96d 100 y 17.5 y
4	Cs-137 Co- 60	661 1332	30.07 y 5.2714 y	0.0762 0.0561	13.714 18.627	3.126 4.476	4.60E-09 5.20E-09	679 861	100 y 17.5 y
5	Ra-226 Sb-122 Cs-137 Eu-152 Pa-234M Co -60	186.1 615 661 723.30 1001.30 1173	1.60E+03 y 2.70 d 30.07 y 13.537 y 6.70 h 5.2714 y	0.133 0.0787 0.0762 0.055 0.0636 0.0587	7.857 13.295 13.714 19.000 16.431 17.802	134.031 3292.5 36.294 8.97 198.42 423.75	3.60E-07 3.60E-10 4.60E-09 4.20E-08 3.80E-10 5.20E-09	372 9145833 7890 213 522157 81490	38.56E+03 y 8.96d 100 y 44.93y 22.24h 17.5 y
6	Th-234 Ra-226 Cs-137 Sb-122 Co-60 Eu-152 Bi-214	92.5 186.1 661 1188 1332 1408.19 1874.42	24.1 d 1.60E+03 y 30.07 y 2.70 d 5.2714 y 13.537 y 0.332 h	0.180 0.133 0.0762 0.0589 0.0561 0.0548 0.0482	5.806 7.857 13.714 17.742 18.627 19.069 21.681	9.003 30.74 4.86 20.85 1,32 3.39 12.15	2.50E-09 3.60E-07 4.60E-09 3.60E-10 5.20E-09 4.20E-08 7.10E-09	3601 85.3 1043 57916 253 80.7 1711	79.99d 38.56E+03 y 100 y 8.96d 17.5 y 44.93y 1.10h
7	Cs-137 Co- 60	661 1173	30.07 y 5.2714 y	0.0762 0.0587	13.714 17.802	5.49 9.48	4.60E-09 5.20E-09	1193 1823	100 y 17.5 y
8	Cs137	661.66	30.07 y	0.0762	13.714	7.521	4.60E-09	1635	100 y
9	Pu- 239 Eu-152 Ra-226 Bi-214 Sb-122 Cs-137 Co -60	77.78 121.78 186.1 609.31 615 661 1173	2.41E+04 y 13.537 y 1.60E+03 y 0.332 h 2.70 d 30.07 y 5.2714 y	0.194 0.159 0.133 0.079 0.0787 0.0762 0.0587	5.837 6.572 7.857 13.228 13.295 13.714 17.802	5.37 11.46 78.39 5.79 2816.4 5268.69 87.45	1.20E-04 4.20E-08 3.60E-07 7.10E-09 3.60E-10 4.60E-09 5.20E-09	0.0475 273 217 815 7823333 11453674 16817	8.00E+04 y 44.93y 38.56E+03 y 1.10h 8.96d 100 y 17.5 y

10	**Pu- 239 Cs-137 Mo-99 Co -60 Eu-152 Bi-214**	**77.78 661 822.97 1173 1408.19 1764.49**	**2.41E+04 y 30.07 y 2.75 d 5.2714 y 13.537 y 0.332 h**	**0.194 0.0762 0.069 0.0587 0.0548 0.0496**	**5.837 13.714 15.145 17.802 19.069 21.056**	**3.23 20.34 3.86 5.85 0.81 0.198**	**1.20E-04 4.60E-09 2.20E-10 5.20E-09 4.20E-08 7.10E-09**	**0.027 4421 17563 1125 19.3 27.8**	**8.00E+04 y 100 y 9.13d 17.5 y 44.93y 1.10h**
11	**Cs137**	**661**	**30.07 y**	**0.0762**	**13.714**	**4.74**	**4.60E-09**	**1030**	**100 y**
12	**Cs137**	**661**	**30.07 y**	**0.0762**	**13.714**	**2.97**	**4.60E-09**	**646**	**100 y**
13	**Cs137**	**661**	**30.07 y**	**0.0762**	**13.714**	**4.14**	**4.60E-09**	**900**	**100 y**
14	**Cs-137 Pr-144**	**661.66 1388.2**	**30.07 y 0.288 h**	**0.0762 0.055**	**13.714 19.000**	**3.633 1673.37**	**4.60E-09 1.80E-11**	**789 92965000**	**100 y 0.79h**
15	**Cs-137 Co -60 Eu-152 Bi-214**	**661.66 1173 1408.01 1764.49**	**30.07 y 5.2714 y 13.537 y 0.332 h**	**0.0762 0.0587 0.0548 0.0496**	**13.714 17.802 19.069 21.056**	**125.67 41.01 6.858 5.192**	**4.60E-09 5.20E-09 4.20E-08 7.10E-09**	**27319 7886 163 731**	**100 y 17.5 y 44.93y 1.10h**
16	**Pu- 239 Zn-65 Bi-214 Cs-137 Co -60 Eu-152 Bi-214**	**77.78 344.95 609.31 661 1173 1408.19 1764.49**	**2.41E+04 y 244 d 0.332 h 30.07 y 5.2714 y 13.537 y 0.332 h**	**0.194 0.101 0.079 0.0762 0.0587 0.0548 0.0496**	**5.837 10.347 13.228 13.714 17.802 19.069 21.056**	**8.25 23144.67 3.519 59.85 70.699 6.39 8.24**	**1.20E-04 2.20E-09 7.10E-09 4.60E-09 5.20E-09 4.20E-08 7.10E-09**	**0.068 10520304 495 13011 13595 152 161**	**8.00E+04 y 809.37d 1.10h 100 y 17.5 y 44.93y 1.10h**
17	**Cs-137 Co-60**	**661.66 1173.24**	**30.07 y 5.2714 y**	**0.0762 0.0587**	**13.714 17.802**	**2.877 89.268**	**4.60E-09 5.20E-09**	**625 17166**	**100 y 17.5 y**
18	**Pa-234M Ra-226 Cs-137 Mo-99 K-40**	**98.5 186.1 661 861.20 1461**	**6.70 h 1.60E+03 y 30.07 y 2.75 d 1.28E+09 y**	**0.175 0.133 0.0762 0.0596 0.0539**	**5.971 7.857 13.714 17.536 19.388**	**820.38 2.61 2.628 52.92 0.696**	**3.80E-10 3.60E-07 4.60E-09 2.20E-10 2.10E-09**	**2158894 7.25 571 240545 331**	**22.24h 38.56E+03 y 100 y 9.13d 4.25E+09 y**

19	**Pa-234M Cs-137 K-40**	**98.5 661 14595.3**	**6.70 h 30.07 y 1.28E+09 y**	**0.175 0.0762 0.0539**	**5.971 13.714 19.388**	**780.912 0.398 0.188**	**3.80E-10 4.60E-09 2.10E-09**	**2055032 86.5 89.5**	**22.24h 100 y 4.25E+09 y**

The present work has illustrated approach to classify the waste for near surface disposal. The new approach is based on to what extent the radionuclide element hazard for human according to its AI values of radionuclide element. The lower value is the higher hazardous element. The ICRP has define the ALI for all radionuclide element for both ingestion and inhalation cases. The study according to the limit cited in table-2 ranking the radionuclide elements into 5 categories, high hazardous HH, low hazardous LH, Very Low Hazards VLH and Extremely Low Hazards ELH. This classification is not considered either the half-life or activity, only the harmful effect of radionuclide is the concern factor. Each class has various radionuclides with wide range of half-life. No common factor is appeared in each class.

Table 2- Classification of radioactive wastes according to AI values

Categories of AI	**Radionuclides**	**AI (Bq)**	**T1/2**	**X cm**	**t**
VHH AI ≤ 0.1Bq	Pu-239	0.0475	2.41E+04 y	5.837	8.00E+04 y
	Pu-239	0.027	2.41E+04 y	5.837	8.00E+04 y
	Pu-239	0.068	2.41E+04 y	5.837	8.00E+04 y
HH AI ≤ 4Bq	U-235	1.44	7.04E+08 y	7.857	23.37E+08 y
LH AI ≤ 10Bq	Ra-226	7.25	1.60E+03 y	7.857	38.56E+03 y
VLH AI ≤ 90Bq	Ra-226	85.3	1.60E+03 y	7.857	38.56E+03 y
	Eu-152	80.7	13.537 y	14.739	44.93y
	K-40	89.5	1.28E+09 y	19.388	4.25E+09 y
	Eu-152	19.3	13.537 y	14.739	44.93y
	Bi-214	27.8	0.332 h	21.681	1.10h
	Cs-137	86.5	30.07 y	13.714	100y
ELH AI >90	Co-60	388	5.2714 y	17.787	17.50 y
	Cs-137	5115	30.07y	13.714	100y
	Eu-152	202	13.537y	14.739	44.93y
	Pa-234m	552829	6.7h	16431	22.24h
	Co-60	21763	5.2714 y	17.787	17.50 y
	Y-88	39695	107d	18.931	355.13d
	Bi-214	173	0.332h	21.069	1.10h
	Pa-234m	2042415	6.7h	16431	22.24h
	Eu-152	154	13.537y	14.739	44.93y
	Sb-122	20046666	2.7d	13.295	8.96d
	Cs-137	3438	30.07y	13.714	100y
	Co-60	4898	5.2714 y	17.787	17.50 y
	Cs-137	679	30.07y	13.714	100y
	Co-60	861	5.2714 y	17.787	17.50 y
	Ra-226	372	1.60E+03 y	7.857	38.56E+03 y
	Sb-122	9145833	2.7d	13.295	8.96d
	Cs-137	7890	30.07y	13.714	100y
	Eu-152	213	13.537y	14.739	44.93y
	Pa-234m	522157	6.7h	16431	22.24h
	Co-60	81490	5.2714 y	17.787	17.50 y
	Th-234	3601	24.1d	5.806	79.99d
	Cs-137	1043	30.07y	13.714	100y
	Sb-122	57916	2.7d	13.295	8.96d
	Co-60	253	5.2714 y	17.787	17.50 y
	Bi-214	1711	0.332	21.681	1.1h
	Cs-137	1193	30.07y	13.714	100y
	Co-60	1823	5.2714 y	17.787	17.50 y

	Cs-137	1635	30.07y	13.714	100y
	Eu-152	273	13.537y	14.739	44.93y
	Ra-226	217	1.60E+03 y	7.857	38.56E+03 y
	Bi-214	815	0.332	21.681	1.1h
	Sb-122	7823333	2.7d	13.295	8.96d
	Cs-137	11453674	30.07y	13.714	100y
	Co-60	16817	5.2714 y	17.787	17.50 y
	Cs-137	4421	30.07y	13.714	100y
	Mo-99	17563	2.75	15.145	9.13d
	Co-60	1125	5.2714 y	17.787	17.50 y
	Cs-137	1030	30.07y	13.714	100y
	Cs-137	646	30.07y	13.714	100y
	Cs-137	900	30.07y	13.714	100y
	Cs-137	789	30.07y	13.714	100y
	Pr-144	92965000	0.288h	19.000	0.79h
	Cs-137	27319	30.07y	13.714	100y
	Co-60	7886	5.2714 y	17.787	17.50 y
	Eu-152	163	13.537y	14.739	44.93y
	Bi-214	731	0.332h	21.681	1.1h
	Zn-65	10520304	244d	10.347	809.37d
	Bi-214	495	0.332h	21.681	1.1h
	Cs-137	13011	30.07y	13.714	100y
	Co-60	13595	5.2714 y	17.787	17.50 y
	Eu-152	152	13.537y	14.739	44.93y
	Bi-214	161	0.332h	21.681	1.1h
	Cs-137	625	30.07y	13.714	100y
	Co-60	17166	5.2714 y	17.787	17.50 y
	Pa-234m	2158894	6.7h	16431	22.24h
	Cs-137	571	30.07y	13.714	100y
	Mo-99	240545	2.75d	15.145	9.13d
	K-40	331	1.28E+09 y	19.388	4.25E+09 y
	Pa-234m	2055032	6.7h	16431	22.24h

The treatment of stored waste that disposal in the tunnels without classification, was done by characterization and classification the radionuclides according to Annual Intake into five categories. And the lower value of Annual Intake is the most dangerous to the public. This means that the more hazards for radionuclide are the lower annual intake value (0.1Bq).

This research suggested method to treatment the radioactive waste by shielding radioactive waste with concrete (density 2.2). The thickness of shielding is the time (t) required to reduce the annual dose to 10% (the last column in the table-2).

Conclusions

The present work we can conclude the following:

1. Radioactive waste has been divided into five categories according to the harmful effect of radionuclides.
2. The limit of annual release in Bq of each nuclide from the disposal site is derived from the annual effective dose values for publics.
3. The suggested method for treatment should be aware that the annual doses of radionuclide buried in specific disposal are within the national dose limit for human.

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