Determination of Alpha Radiation Dose to Skin Due to Artificial and Natural Radionuclides from the Deposition of Different Material Samples

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Abstract

Artificial alpha-emitting radionuclides are significantly utilized in nuclear medicine. So, the skin of practitioners as well as patients in nuclear medicine and workers in the industry of radiopharmaceuticals may be accidentally contaminated even for a very short time by these radionuclides. Natural alphaemitting radioisotopes such as those belonging to the uranium and thorium series are deposited on the human skin from the application of various natural material samples. The mean absorbed dose in skin due alpha-particles emitted by natural and artificial radionuclides was calculated by using a Monte Carlo computer code based on determining residual energies of the emitted alphaparticles. Alpha-activities were determined for radiopharmaceutical and natural material samples. A new dosimetric model based on exploiting data obtained for alpha-activities and mean alpha absorbed doses due to artificial and natural material samples for evaluating committed equivalent doses to skin of workers was developed. The influence of the application time, alphadisintegration ratio, half-life of the radionuclide, and contaminated skin surface on committed equivalent dose was investigated. Maximum values of the committed equivalent dose to skin were found equal to 0.552 mSv cm⁻² y⁻¹ for alpha-particles emitted by artificial radionuclides and $0.16 \text{ mSv cm}^{-2} \text{ y}^{-1}$ for those emitted by ²³⁸U, ²³²Th and ²²²Rn natural radioisotopes, respectively.

Keywords: Artificial and natural alpha-emitting radionuclides; Monte Carlo simulation; Radiation dose assessment to skin

1. INTRODUCTION

With a surface area of about 2 m² and a weight of about 2.1 kg, the skin is the largest organ of the human body. It protects individuals from the hazards of the environment, regulates body temperature and limits sensation of touch, heat and cold. The skin is composed of a series of three layers: The epidermis, dermis and hypodermis. The critical cells in the skin are in the basal layer of the epidermis [1]. Alpha-particles are helium nuclei made up of two protons and two neutrons which have short range and high linear energy transfer. Alpha-particle emitting radionuclides are of great interest in alpha-radioimmunotherapy and targeted therapy for the treatment of malignant diseases [2-7]. Patients and practitioners in nuclear medicine as well as workers in the industry of radiopharmaceuticals may be accidentally contaminated by alpha-emitting radiopharmaceuticals. Natural radioisotopes belonging to the ²³⁸U and ²³²Th series are deposited on the skin of individuals from the application of various material samples such as building materials [8], olive oil [9], black soap [10], thermal waters [11], and medical drugs [12]. It is then necessary to assess alpha-radiation dose to the skin of workers to avoid any radiation dose enhancement.

In the present work, mean absorbed dose in skin was calculated by using a Monte Carlo computer code based on generating uniformly alpha-particles in a material layer deposited on skin and determining the residual energy of these particles. Alpha-activities of the deposited material samples were evaluated. Committed equivalent doses to skin due to artificial and natural alpha-emitting radionuclides from the application of different material samples were determined.

2. MATERIAL AND METHODS

a. Calculation of the mean absorbed dose to skin due to alpha-particles

The epidermis of the human skin is divided into several clearly defined zones [13]. Indeed, when a material layer of 1 mm depth is placed on the skin of an individual (Fig. 1), the emitted alpha-particles have ranges of several tens of microns (20 to 100μ m). This is comparable with the depth of the basal layer of the epidermis which is more sensitive (50 to 100μ m) [14].

An alpha-particle of index j and initial energy E_{α_j} emitted from a nucleus localized on the point P inside a material layer (Fig. 1) has a range:

$$\overline{PF} = x_i + R_i^{skin} \tag{1}$$

where x_j ($x_j \le R_j, R_j$ is the range of the alpha-particle inside the material layer) is the distance between the emission point and the skin surface (Fig. 1) and R_j^{skin} is the range of the alpha-particle in skin. The ranges of alpha-particles in the material layer and skin were calculated using a TRIM programme [15].



Fig. 1. Ranges of an alpha-particle inside a material layer ($\overline{PI} = x_j$) and epidermis ($\overline{IF} = R_j^{skin}$). $E_{\alpha j}$ is the initial alpha-particle energy and $E_{\alpha j}^{Res}$ its residual energy on the point I. The deposited material layer has a depth of about 1mm.

The alpha-particle residual energy $E_{\alpha_j}^{\text{Res}}$ which corresponds to the $(R_j - x_j)$ range is determined by using the energy-range relation in the material sample (an example is given in Fig. 2a for alpha-particles in a radiopharmaceutical sample). By using the energy-range relation in skin one can determine the range of the alpha-particle in skin R_j^{skin} (Fig. 2b). For $x_j = R_j$, $E_{\alpha_j}^{\text{Res}} = 0 \text{ MeV}$; there is no energy loss of alpha-particles in skin (case 1 of Fig. 1). For $x_j = 0 \ \mu m$, $E_{\alpha_j}^{\text{Res}} = E_{\alpha_j}$; the energy loss of alphaparticles in skin is maximum (R_j^{skin} maximum) (case 3 of Fig. 1). For $x_j \prec R_j$, $E_{\alpha_j}^{\text{Res}} \prec E_{\alpha_j}$; the ranges of alpha-particles in skin are lower than those corresponding to $x_j = 0 \ \mu m$ (case 2 of Fig. 1).

(4)

Let us consider a cylindrical material layer of 1 cm² basis surface and 1 mm depth deposited on the skin surface of an individual as shown in Fig. 3. An alpha-particle of index j and initial energy E_{α_i} generated at point P (Fig. 3) at a distance x_j from the

skin which reaches the skin has a range R_i^{skin} in skin. Since the deposited material layer has a depth of 1 mm which is greater than the radon diffusion length in the considered materials, alpha-particles emitted by radon in ambient air cannot reach the skin of individuals (Fig. 1).

The distance x_i is given by (Fig. 3):

$$x_j = \frac{t}{\cos\theta} \text{ if } \qquad q > \overline{O_1'Q_1}$$
 (2)

where

$$\overline{0_1'Q_1}^2 = r^2 + t^2 tg^2(\theta) + 2r ttg(\theta) \cos(\psi)$$
(3)

q is the radius of the cylindrical material layer.

The calculation of the absorbed dose in skin due to the emitted alpha-particles consists firstly on generating random numbers by using a programme called random subroutine, based on a congruential method [16], and calculating the x_i distances and, R_j and R_i^{skin} ranges by using a programme called AMADS (Alpha Mean Absorbed Dose in Skin) which is represented schematically in Fig. 4.

The uniform random sampling of the emission point P and emission direction is achieved by computing the distance from the center axis r, the depth t, $\cos \theta$ (θ between 0 and $\frac{\pi}{2}$) and Ψ (Fig. 3) with four uniform random numbers [16-18]: $r = q_{\sqrt{\xi_1}} \qquad , 0 \le \xi_1 < 1$ $t = R_j \xi_2 \qquad , 0 \le \xi_2 < 1$ $\cos\!\theta \!=\! \xi_3 \qquad , 0 \!\leq\! \xi_3 \!<\! 1$

$$\psi = 2\pi\xi_4$$
 , $0 \le \xi_4 < 1$

where R_j is the range of an alpha-particle of index j and initial energy E_{α_j} in the material sample.

The absorbed dose (Gy) in a cylinder of 1 cm^2 basis surface and $R_j^{skin} \cos \theta$ depth of skin due to an alpha-particle of index j and residual energy $E_{\alpha_j}^{\text{Res}}$ is:

$$D_{j} = \frac{k E_{\alpha j}^{Res}}{m}$$
(5)

where $m = d_{Skin}R_j^{Skin} \cos \theta x 1 \text{cm}^2$ is the mass of the cylindrical volume of skin, d_{Skin} is the density of skin and $k = 1.6 \times 10^{-13} \text{ J MeV}^{-1}$ is a conversion factor.

Eq. (5) can be rewritten:

$$D_{j} = \frac{k E_{\alpha j}^{\text{Res}}}{d_{\text{Skin}} R_{j}^{\text{Skin}} \cos \theta \, x \, 1 \, \text{cm}^{2}}$$
(6)

For a large number N_r^j of α -particles of index j reaching the surface skin with a residual energy $E_{\alpha_i}^{\text{Res}}$ the mean absorbed dose in skin is:

$$\left< D_{j}(skin) \right> = \frac{\sum D_{j}}{N_{r}^{j}}$$
(7)

An alpha-particle of index j and initial energy E_{α_j} reaches the basal layer of the human skin when:

$$x_j < R_j \text{ and } R_j^{\text{Skin}} > (50 \mu \text{m/cos}\theta)$$
 (8)

The mean absorbed dose in the basal layer of an individual is given by:

$$\left\langle D_{j}(BL)\right\rangle = \frac{\sum D_{j}}{N_{BL}^{j}} \tag{9}$$

where N_{BL}^{j} is the number of alpha-particles reaching the basal layer of skin with a residual energy $E_{\alpha_{i}}^{\text{Res}}(BL)$.

The mean absorbed dose per fluence in the basal layer of the epidermis is given by:

$$\frac{\left\langle \mathbf{D}_{j}(\mathbf{BL})\right\rangle}{\phi} = \frac{\sum \mathbf{D}_{j}}{\mathbf{N}_{\mathrm{BL}}^{j}} \times 1 \,\mathrm{cm}^{2}$$
(10)

b. Committed equivalent dose to skin due to alpha-particles from the deposition of different material samples

Alpha-equivalent dose rates (Sv s^{-1}) to the skin of individuals due to an alpha-particle emitted by a radionuclide j from the application of a material sample is given by:

$$H_{skin}(j)(t) = \langle D_j(Skin) \rangle K_j \quad A_C^{skin}(j)(t) W_R$$
(11)

where $A_c^{skin}(j)(t)$ (Bq) is the alpha-activity, at time t, in skin due to a radionuclide j,W_R is the radiation weighting factor which is equal to 20 for alpha-particles [13], and K_j is the alpha-disintegration branching ratio.

The $A_c^{skin}(j)(t)$ alpha-activity is given by:

$$\mathbf{A}_{c}^{skin}(j)(t) = \frac{1}{2} \mathbf{A}_{c}^{sample}(j) \mathbf{e}^{-\lambda_{j}t} \mathbf{x} \ 1 \, \mathrm{cm}^{2} \, \mathbf{x} \, R_{j}$$
(12)

where $A_c^{sample}(j)$ in Bq cm⁻³ is the alpha-activity due to a radionuclide j inside a material sample, λ_j is the radioactive decay constant of a radionuclide j and R_j (in cm) is the range of an alpha-particle of index j in the material sample. The term $\frac{1}{2}$ means that only half of the emitted alpha-particles inside a material sample may lose their energies inside the skin.

By integrating Eq. (11), committed equivalent dose (Sv) to skin due to an alphaparticle of residual energy $E_{\alpha_j}^{\text{Res}}$ emitted by a radionuclide j from the application of a material sample is given by:

$$H_{skin}(j) = \frac{\langle D_{j}(Skin) \rangle K_{j} W_{R}}{2\lambda_{j}} A_{C}^{sample}(j) (1 - e^{-\lambda_{j}} t_{a}) x 1 cm^{2} x R_{j}$$
(13)

where t_a is the application time of the material sample on skin.

Similarly committed equivalent dose to the basal layer of the epidermis from the application of a material sample is given by:

$$H_{BL}(j) = \frac{\langle D_{j}(BL) \rangle K_{j} W_{R}}{2\lambda_{j}} A_{C}^{sample}(j) \left(1 - e^{-\lambda_{j} t_{a}}\right) x \ 1 \text{ cm}^{2} x R_{j}$$
(14)



Fig. 2. Alpha-particle range-energy relationship for a material sample (a) and skin (b).



Fig. 3. Arrangement of a cylindrical material layer of 1 cm² surface and 1 mm depth deposited on the skin surface of an individual. q is the radius of the material layer. $\overline{PI} = x_j$



Fig. 4. Flow chart of the AMADS Fortran programme which is used to calculate the mean absorbed dose in skin of individuals from deposition of alpha-emitting radionuclides. We employ the same symbols as in the text. It is therefore self-explanatory. $N_0 = 10^6$ alpha-particles.



Fig. 5. Variation of the mean absorbed dose in the corneum, granulosum and spinosum strata (skin) and basal layer of the epidermis as a function of alpha-particle initial energy.

RESULTS AND DISCUSSION

a. Mean absorbed dose to skin from the deposition of alpha-emitting radionuclides

Mean absorbed dose in skin due to alpha-particles generated in a material layer deposited on the skin of an individual was evaluated by using the AMADS (Alpha mean absorbed dose in skin) Monte Carlo code. Data obtained are represented in Fig. 5. The statistical uncertainty of the mean absorbed dose determination is of 0.5 %. It is to be noted that the mean absorbed dose in skin increases, reaches a maximum at $E_{\alpha_j} = 3$ MeV and decreases when E_{α_j} increases. This is due to the fact that the stopping power of skin for the incident alpha-particles ($E_{\alpha_j}^{\text{Res}}/R_j^{skin}$) (Eq. (6)) increases in the [0-3MeV] energy interval and then decreases for energies larger than 3 MeV. One can also note that only alpha-particles of initial energy greater than 6.75 MeV can reach the sensitive basal layer of the epidermis. Mean absorbed dose in skin was calculated for alpha-particles emitted by different artificial radionuclides used in nuclear medicine. Data obtained are shown in Table 1. Only alpha-particles emitted by ²¹²Po can reach the basal layer of the epidermis (Table 1). Mean absorbed dose was calculated for different contaminated skin surfaces belonging to the [1-50 cm²]

interval from accidental application of various radiopharmaceuticals. An example is given in figure 6 for the ¹⁴⁹Tb and ²²⁵Ac radionuclides. It is to be noted that the mean absorbed dose decreases when the contaminated skin surface increases.

Table 1. Data obtained for mean absorbed dose in the corneum, granulosum and spinosum strata ($\langle D_j(skin) \rangle$) and basal layer ($\langle D_j(BL) \rangle$) of the epidermis from the application of different alpha-emitting radionuclides.

Radionuclide (j)	Eαj (MeV)	Branching ratio (%) λ_{j} (s ⁻¹)		⟨D _j (skin)⟩ (nGy)	$\langle D_j(BL) \rangle$ (nGy)
²²⁴ Ra	5.68	100	2.2 x 10 ⁻⁶	488.2±0.7	0
²¹² Bi	6.05	35.94	190.63 x 10 ⁻⁶	479.8±0.7	0
²¹³ Bi	5.87	2	253.34 x 10 ⁻⁶	483.9±0.7	0
²²⁵ Ac	5.73	100	0.80 x 10 ⁻⁶	487.1±0.7	0
²¹¹ At	5.87	41.80	26.74 x 10 ⁻⁶	483.9±0.7	0
²³⁰ U	5.80	100	0.386 x 10 ⁻⁶	485.5±0.7	0
²¹⁰ Po	5.30	100	0.058 x 10 ⁻⁶	496.8±0.7	0
²²³ Ra	5.979	100	0.704 x 10 ⁻⁶	481.4±0.7	0
²¹² Po	8.78	100	2310490	423.1±0.6	300±1
¹⁴⁹ Tb	4.00	22.6	46.961 x 10 ⁻⁶	523.5±0.8	0



(a) Terbium 149



Fig. 6. Variation of the mean absorbed dose in skin as a function of skin contaminated surface from accidental application of ¹⁴⁹Tb (a) and ²²⁵Ac (b) radionuclides.

Mean absorbed doses in skin due to natural radionuclides (²³⁸U, ²³²Th and ²²²Rn) from the application of various building material samples were determined. Data obtained are shown in Table 2.

Table 2. Data obtained for the mean absorbed dose in skin ($\langle D_j(skin) \rangle$) due to ²³⁸U, ²³²Th and ²²²Rn from the application of different building material samples by workers.

Material		<dj(skin)> (10⁻⁷Gy)</dj(skin)>	
sample	238U	²³² Th	²²² Rn
Cement	5.22±0.01	5.25±0.01	4.87±0.01
Marble	5.23±0.01	5.26±0.01	4.96±0.01
Plaster	5.21±0.01	5.24±0.01	4.95±0.01
Clay	5.23±0.01	5.26±0.01	4.97±0.01

In order to validate our calculation method, alpha-particles reaching the surface skin with residual energies given in Annex G of the ICRP Publication 116 [14] and normally incident on skin have been considered. Data obtained by the two methods for the absorbed dose per fluence to the basal layer of skin were found in good agreement with each other (Table 3).

E_{α_i}	Mean absorbed dose per fluence (µGy.cm ²)				
(MeV)	This method	ICRP dose coefficient			
6.5	-	0.00111			
6.8	0.0201	0.0256			
7.0	0.0403	0.0420			
7.5	0.0697	0.0752			
8.0	0.098	0.103			
8.5	0.109	0.128			
9.0	0.142	0.150			
9.5	0.136	0.140			
10.0	0.173	0.180			

Table 3. Values of mean absorbed dose per fluence in the basal layer of the epidermis obtained by this method and those given by the ICRP [14], for mono-energetic alphaparticles normally incident on skin.

b. Alpha committed equivalent dose to skin from the application of different material samples

When injecting radiopharmaceutical solutions containing alpha-emitting nuclei to patients some drops may be deposited on their skin. Workers in radiopharmaceutical industry may also be contaminated by alpha-emitting radioisotopes. Committed skin determined different equivalent dose to was for alpha-emitting radiopharmaceuticals from accidental application during 5, 10 and 20 minutes per year for a deposited activity of 20 Bq cm⁻³ (20 drops). Data obtained are shown in Table 4. It is to be noted that committed equivalent dose to skin increases with the application time of radiopharmaceuticals. Even if ²¹²Po emits alpha-particles with energy higher than those emitted by the other radionuclides, committed equivalent dose to skin due this radionuclide is negligible compared with those due to the other radionuclides. This is because ²¹²Po has a half-life lower than those of the other radionuclides (Table 1). One can note that committed equivalent dose due to ²¹³Bi is

clearly lower than those due to the other radionuclides. This is due to the fact that 213 Bi has a lower alpha disintegration branching ratio (2%) than the other radionuclides. It is obvious, according to Eq. (13) that committed equivalent dose to skin due to a radionuclide j increases when the corresponding deposited activity increases. The maximum committed equivalent dose to skin was found equal to 0.552 mSv cm⁻² y⁻¹, obtained for accidental application of ²²³Ra during 20 minutes per year for a deposited activity of 20 Bq cm⁻³ (Table 4), which is smaller than the dose limit for workers which is of 500 mSv cm⁻² y⁻¹[1].

Table 4. Data obtained for the committed equivalent dose to the corneum, granulosum and spinosum strata ($H_{skin}(j)$) and basal layer ($H_{BL}(j)$) of the epidermis from accidental application, during 5, 10 and 20 minutes, of various radiopharmaceuticals. We considered a volume of 20 drops placed on the skin of individuals corresponding to an activity of 20 Bq/cm³. The relative uncertainty is of 0.5 %.

Radionuclide	H _{Skin} (j)	H _{Skin} (j)	H _{Skin} (j)	H _{BL} (j)	H _{BL} (j)	H _{BL} (j)
(j)	$(\mu Sv/cm^2/y)$					
	t _A =5 min	t _A =10 min	t _A =20 min	t _A =5 min	t _A =10 min	t _A =20 min
²²⁴ Ra	129.4±0.2	258.7±0.4	517.02±1.03	0	0	0
²¹² Bi	49.0±0.1	95.2±0.2	180.19±0.36	0	0	0
²¹³ Bi	2.60±0.01	5.00±0.01	9.31±0.02	0	0	0
²²⁵ Ac	130.8±0.3	261.7±0.5	523.26±1.05	0	0	0
²¹¹ At	56.2±0.1	111.9±0.2	222.07±0.44	0	0	0
²³⁰ U	133.0±0.3	265.8±0.5	531.48±1.06	0	0	0
²¹⁰ Po	118.5±0.2	236.9±0.5	473.89±0.95	0	0	0
²²³ Ra	138.1±0.3	276.2±0.5	552.34±1.10	0	0	0
¹⁴⁹ Tb	18.49±0.04	36.72±0.05	72.42±0.14	0	0	0
²¹² Po	(3.23±0.01)10 ⁻⁷	(3.23±0.01)10 ⁻⁷	(3.23±0.01)10 ⁻⁷	(2.29±0.01)10 ⁻⁷	(2.29±0.01)10 ⁻⁷	(2.29±0.01)10 ⁻⁷

Alpha-activities per unit volume due to ²³⁸U, ²³²Th and ²²²Rn were measured inside different building material samples by using a method based on using CR-39 and LR-

115 II solid state nuclear track detectors (SSNTDs) [19]. Data obtained are shown in Table 5. The uncertainty of the ²³⁸U, ²³²Th and ²²²Rn concentrations determination is of the order of 8 %. Committed equivalent doses to the skin of individuals due to ²³⁸U, ²³²Th and ²²²Rn from the application of different building material samples by workers were evaluated. Data obtained are shown in Table 5. The uncertainty of the committed equivalent doses determination is of 9 %. It is to be noted that committed equivalent doses to skin due to ²³⁸U, ²³²Th and ²²²Rn from the application of clay are clearly higher than those resulting from the application of the other building material samples. This is because clay shows higher ²³⁸U, ²³²Th and ²²²Rn alpha-activities than the other building material samples. One can also note that committed equivalent dose to skin due to ²²²Rn is clearly lower than those due to ²³⁸U and ²³²Th from the application of the studied building material samples. This is due to the fact that ²²²Rn has a lower half-life (3.82 d) than those of ²³⁸U and ²³²Th (Eq. (13)). A maximum value of the total committed equivalent dose to skin due to ²³⁸U, ²³²Th and ²²²Rn was found equal to 0.16 mSv cm⁻² y⁻¹, obtained from the application of clay by workers (2016 hours per year) (Table 5), which is smaller than the dose limit for workers which is of 500 mSy cm⁻² v^{-1} [1].

Table 5. Table 5. Committed equivalent doses to the epidermis of skin due to 238 U, 232 Th and 222 Rn from the application of different building material samples.

Building material sample (application time)	A _c (²³⁸ U) (Bq.m ⁻³)	A _c (²³² Th) (Bq.m ⁻³)	A _c (²²² Rn) (Bq.m ⁻³)	$H_{Skin}(^{238}U)$ (µSv.cm ⁻² .y ⁻¹)	$H_{Skin}(^{232}Th)$ (µSv.cm ⁻² .y ⁻¹)	$H_{Skin}(^{222}Rn)$ (µSv.cm ⁻² .y ⁻¹)
Cement (8h/day)	165±10	165±10	165±10	7.90± 0.48	7.48±0.45	0.78±0.05
Marble (8h/day)	235±14	205±13	235±12	13.78± 0.82	11.39±0.72	1.26±0.06
Plaster (8h/day)	82±5	53±4	82±5	11.36±0.69	6.95±0.52	1.03±0.06
Clay (8h/day)	998±60	647±39	998±60	91.86±5.52	56.42±3.40	8.42±0.51

4. CONCLUSION

It has been shown by this study that by using a Monte Carlo computer code, one can determine mean absorbed dose in skin due to alpha-emitting radionuclides from the application of various artificial and natural material samples. A new dosimetric model was described for determining committed equivalent dose to skin due to alpha-

emitting nuclei from the application of various radiopharmaceutical and building material samples. It is concluded that committed equivalent dose to skin is influenced by the disintegration branching ratio, half-life of the radionuclide, deposited activity, contaminated skin surface and application time of the material samples. Therefore, medical personnel in hospitals as well as workers in the radiopharmaceutical industry should avoid any contamination by alpha-emitting radionuclides when handling radiopharmaceuticals. Also workers in building material factories should protect their skin from building material dusts to avoid any enhancement of exposure to alpha radiation. This dosimetric method is a good tool for assessing alpha radiation doses due to the cutaneous application of diverse products such as creams, medical drugs, plant oils...

5. ACKNOWLEDGMENTS

This work was realized under an URAC-15 research contract with the CNRST, Rabat, Morocco.

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