

## THE COUNCIL

Having regard to Article 5(a) and (b) of the Convention on the Organisation for Economic Co-operation and Development of 14th December 1960 ;

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1. At its 132nd Meeting on 24th January 1967 the Council AGREED that, in accordance with Article 6(3) of the Convention and Rule 19(b) of the Rules of Procedure, pending its ratification by Germany, this Decision should apply provisionally between other Member countries.

Considering that the European Nuclear Energy Agency is charged with encouraging the elaboration and harmonisation of legislation relating to nuclear energy in participating countries, in particular with regard to the protection of public health ;

Having regard to the Decision of the Council of 18th December 1962 on the Adoption of Radiation Protection Norms [Doc. No. C(62)187(Final)] ;

Considering that in adopting these Norms the Council decided that necessary measures should be taken to ensure that adequate protection against the hazards of ionizing radiations for the population is provided and maintained wherever radioactive materials are possessed or used ;

Considering that ionizing radiations are produced by radioluminous paint used on the components of certain timepieces ;

Considering the importance of establishing special standards to ensure that the users of radioluminous timepieces and the whole population are adequately protected, and with a view toward facilitating international trade in this field ;

## I. DECIDES

The Governments of Member countries shall ensure that appropriate measures are taken to provide adequate protection against the hazards of ionizing radiations for users of radioluminous timepieces and for the whole population.

II. RECOMMENDS to the Governments of Member countries that the measures referred to above be based upon the Radiation Protection Standards for Radioluminous Timepieces annexed hereto and that the control procedures described in the Appendix to these Standards be used to ensure that the manufacture of radioluminous timepieces complies with the requirements laid down in these Standards.

III. DECIDES that the Radiation Protection Standards for Radioluminous Timepieces (including their Appendix) will be published, as well as Annexes to these Standards which provide background information on the technical and health aspects on which the Standards are based.

## RADIATION PROTECTION STANDARDS FOR RADIOLUMINOUS TIMEPIECES

Recommendations  
drawn up by a joint group of experts  
of ENEA and I.A.E.A.

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### *Introduction*

In preparing the Draft Recommendations set out below, the European Nuclear Energy Agency (ENEA) and the International Atomic Energy Agency (I.A.E.A.), hereinafter referred to as «the Agencies», took into account national regulations and recommendations at present in force or under consideration, as well as scientific data related to the use of timepieces containing components bearing radioactive luminous paint.

The work has been undertaken with a view to obtaining international agreement on recommendations to promote safety, in compliance with internationally agreed radiation protection standards, and to facilitate international trade.

The Agencies were assisted in this work by a group of experts who met in February 1964 and in May 1965 at the ENEA Headquarters in Paris. A first Draft Standards was drawn up as a result of the work of the group of experts and was circulated in Member countries of both organisations for consideration and comment.

This document, which is the final version of the Draft Standards, was finalized within the ENEA Health and Safety Committee in May 1966, after a general discussion in the light of the comments submitted by various Member countries and the International Commission on Radiological Protection (ICRP).

The composition of the group of experts who took part in the drawing up of the Draft Standards was as follows :

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#### 1. *Explanation of Terms Used*

For the purposes of these Standards, the following explanations of terms used are given :

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| (1) <i>Ordinary timepieces</i>           | Wrist or pocket watches or clocks, which contain a quantity of radioactive material acceptable for ordinary use.                                       |
| (a) Wristwatches                         | Under normal conditions of use, wristwatches are worn on the wrist.  |
| (b) Pocket watches                       | Watches worn on the individual, other than on the wrist.   |
| (c) Clocks                               | Timepieces which are not worn on the individual.   |
| (2) <i>Special timepieces</i>            | Timepieces usually worn by the individual, containing quantities of radioactive material to produce greater luminosity necessary for special purposes. |
| (3) <i>Total activity of a timepiece</i> | The total activity of the radioluminous paint present in a timepiece, regardless of which parts are luminised.   |



import/export and disposal of timepieces containing components bearing radioluminous paint. Such operations shall be carried out in accordance with national and international Norms for Radiation Protection.

- 2.4 These Standards are not intended to cover radiation protection aspects for special timepieces containing quantities of radioactive material which exceed the limits set out in these Standards ; these shall be subject to the jurisdiction of the appropriate national authority.
- 2.5 These Standards are not intended to cover radiation protection aspects relevant to workers in the radioluminous paint and watch industry <sup>(1)</sup>. Nor are they intended to cover watchmakers who repair and maintain radioluminous timepieces, who should be informed of the potential hazards involved and should observe appropriate protective measures.

### 3. *General Requirements*

- 3.1 Selection of radionuclides for use in luminous paints, and limitation of activity levels for dials and hands of timepieces shall be governed by consideration of :

- (i) the quantity of a radionuclide necessary to obtain the required luminosity of the dials and hands of a timepiece ;
- (ii) the external radiation doses normally received by the user from the activity referred to under (i) ;
- (iii) the doses which would result from internal radiation if particles of the radioactive luminous paint were ingested or inhaled, with a view to minimising exposure from external radiation and to keeping the potential risk of exposure from internal radiation as low as possible, for both individual members of the public and the whole population.

- 3.2 Apart from the considerations referred to in 3.1, the quantity of radioactive material in a timepiece shall be governed by the consideration of whether it is for ordinary or special use. Timepieces shall therefore be classified as :

- (a) Timepieces for ordinary use (ordinary timepieces) ;
- (b) Timepieces for special use (special timepieces).

Ordinary timepieces shall be free for general distribution, which will be wide and uncontrolled. Consequently the quantity of radioactive material they contain must be limited to amounts that will satisfy the aims of these standards as set out in paragraph 2.2.

Special timepieces are designed for uses for which greater luminosity than is necessary for ordinary timepieces is required. Their distribution is limited and their use is, in general, restricted to a limited period of time. Therefore, the quantity of radioactive material in special timepieces can be greater than is acceptable for ordinary timepieces and still satisfy the aims of these standards as set out in paragraph 2.2.

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1. These aspects have been dealt with in the I.L.O. « Manual of Industrial Radiation Protection, Part V. Guide on Protection against Ionizing Radiation in the Application of Luminous Compounds », Geneva, 1964.

3.3 The watch casing (case and transparent cover) shall be such that under normal conditions of use :

- (i) the user is protected against direct contact with radioactive dials and hands ;
- (ii) low-energy beta-radiation is effectively absorbed by the watch casing.

3.4 The adhesion of radioluminous paint to dials and hands of timepieces shall be such that, under normal conditions of use, it remains fixed on these components.

#### 4. *Special Requirements*

4.1 Radionuclides permitted for use in luminous paint for dials and hands of timepieces<sup>1</sup>.

4.1.1 Subject to 4.1.3, the use of radionuclides in luminous paint for dials and hands of ordinary and special timepieces shall be restricted to :

- (i) Tritium (H-3)
- (ii) Promethium-147 (Pm-147)
- (iii) Radium-226 (Ra-226)

However, the use of H-3 and Pm-147 should be preferred as far as possible.

Ra-226 shall not be used for pocket watches, regardless of whether these are ordinary or special timepieces.

4.1.2 The luminous paint used for the dials and hands of a timepiece should be activated with one and the same radionuclide<sup>2</sup>.

4.1.3. Any other radionuclide considered for use in timepieces shall be subject to a separate decision by the appropriate national authority based on the general requirements set out in paragraphs 3.1 - 3.4<sup>3</sup>. The appropriate national authority shall inform the international organisations concerned of such decisions.

4.2 Total activity of radionuclides permitted per timepiece<sup>4</sup>.

4.2.1 *Ordinary timepieces*

For ordinary timepieces, the total activity of radionuclides permitted according to 4.1.1 shall be limited as follows :

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$$\left( \frac{X}{L_X} + \frac{Y}{L_Y} + \dots \right) \leq 1$$

X, Y represent the activity of the radionuclide actually used,  
 $L_X, L_Y$  represent the respective maximum permissible total activity.

3. The Agencies are prepared to consider the amendment of the list of radionuclides specified for use at a later stage, when new scientific information is available.

4. See Annex II.

a) *Wristwatches*

| Radionuclide | Average      | Maximum       |
|--------------|--------------|---------------|
| H-3          | 5 mCi        | 7.5 mCi       |
| Pm-147       | 100 $\mu$ Ci | 150 $\mu$ Ci  |
| Ra-226       | 0.1 $\mu$ Ci | 0.15 $\mu$ Ci |

b) *Pocket Watches*

| Radionuclide | Average      | Maximum      |
|--------------|--------------|--------------|
| H-3          | 5 mCi        | 7.5 mCi      |
| Pm-147       | 100 $\mu$ Ci | 150 $\mu$ Ci |

c) *Clocks*

| Radionuclide | Average       | Maximum      |
|--------------|---------------|--------------|
| H-3          | 7.5 mCi       | 10 mCi       |
| Pm-147       | 150 $\mu$ Ci  | 200 $\mu$ Ci |
| Ra-226       | 0.15 $\mu$ Ci | 0.2 $\mu$ Ci |

4.2.2. *Special timepieces*

For special timepieces, the total activity of radionuclides permitted according to 4.1.1 shall be limited as follows :

| Radionuclide        | Maximum Total Activity |
|---------------------|------------------------|
| H-3                 | 25 mCi                 |
| Pm-147              | 0.5 mCi                |
| Ra-226 <sup>1</sup> | 1.5 $\mu$ Ci           |

## 4.3 Adhesion of radioluminous paint on dials and hands.

4.3.1 The degree of adhesion of radioluminous paint on dials and hands should be subject to tests. The adhesive agent of the paint should provide adhesion on the painted surfaces in a manner which will withstand bending, vibrating and changes of temperature normally incident to the use of timepieces.

4.3.2 The radioactive material should be bound in the luminous compound in a not readily soluble form, either by means of its chemical composition or with the help of a binding agent.

## 4.4 Casing

Timepieces with dials and/or hands bearing radioluminous paint shall be enclosed in a case, the transparent cover of which must have a thickness equivalent to not less than 50 mg/cm<sup>2</sup> at any point. The

1. Not permitted for pocket watches (see paragraph 4.1.1).



casing and the transparent cover should be strong enough to withstand conditions normally incident to use and conditions incident to minor accidents.

#### 4.5 Marking

##### 4.5.1 Ordinary timepieces

Ordinary timepieces do not require a special marking.

##### 4.5.2. Special timepieces

Special timepieces must bear a specific marking on the dial to advise the watchmaker and the user of their special nature, and to permit the appropriate national authority to take measures for the introduction of adequate systems of notification, declaration, registration and/or licensing if so deemed necessary.

The marking should be as follows :

| Radionuclide   | Marking    | To indicate upper limit of activity as |
|----------------|------------|--|
| Tritium        | « T 25 »   | 25 mCi H-3                             |
| Promethium-147 | « Pm 0.5 » | 0.5 mCi Pm-147                         |
| Radium-226     | « Ra 1.5 » | 1.5 mCi Ra-226                         |

#### 4.6 Tests

Special requirements as set out in paragraphs 4.1 to 4.5 shall be subject to tests ; these shall be carried out under the jurisdiction of the appropriate national authority. Recommendations for control procedures for radioluminous timepieces are given in the Appendix.

#### 4.7 Exemption from notification, registration and/or licensing.

Provided that the detailed requirements listed above are met, the use of ordinary watches shall be exempted from notification, declaration, registration and/or licensing. Unless decided otherwise by the appropriate national authority, the exemption from notification, declaration, registration and/or licensing is extended to special timepieces containing Tritium, but these requirements may be advisable in the case of Promethium-147 and in particular of Radium-226.

## APPENDIX

### *Control procedures for radioluminous timepieces*

#### 1. Introduction

1.1 Tests are necessary in order to ensure that the special requirements laid down in the Standards for Radioluminous Timepieces are met. By these Standards, it is necessary to ensure that :

- (i) The use of radionuclides in luminous paint for dials and hands of timepieces is restricted to selected radionuclides (H-3, Pm-147 or Ra-226) ;
- (ii) The total activity per timepiece is limited ;
- (iii) Marking of dials is applied if the total activity of a timepiece exceeds the activity levels set out for ordinary timepieces ;
- (iv) The adhesion of radioluminous paint to dials and hands of timepieces shall be such that, under normal conditions of use, it remains fixed on these components.
- (v) The thickness of the casing is not lowered below minimum thickness values.

The aim of the recommended tests is therefore to ensure compliance with the provisions of the Radiation Protection Standards. They do not relate to the quality of timepieces or their components.

- 1.2 It is not practicable to include in a recommendation of an international character, tests of all the properties of luminous paints which it might be of interest to examine. A minimum control programme is therefore proposed in order to determine compliance with the requirements as previously listed.

The tests may be carried out on the components or, if possible, on the complete timepiece. Besides the minimum control programme, it may be advisable, however, that producers perform additional quality tests on the paints or painted hands and dials, before they are used in the production of timepieces.

- 1.3 The control procedure should be carried out under the jurisdiction of the appropriate national authority and should comprise both prototype tests and quality control.

There may be differences in the organisation of control procedures from country to country, depending on different existing regulations. Details of the organisation of the control are therefore left to be decided by the appropriate national authority, but it is recommended that it should be conducted on a common basis according to the recommendations given in the Minimum Control Programme below.

- 1.4 Exemption from these regulations may be granted by the appropriate authority if the producer can guarantee that the requirements in the Minimum Control Programme have been fulfilled.

## 2. *Minimum Control Programme*

### 2.1 *Explanation of terms*

- (a) *Prototype tests* are tests to determine that the combination of the radioluminous paint, materials of construction and methods of manufacture are such that the timepiece or its components meet the requirements of the Standards.
- (b) *Quality control* comprises tests to ensure that the paint, components and manufacturing methods are the same as those used to produce the prototype timepiece or component, and the quality of the product is the same as the quality of the timepiece or component on which the prototype tests were conducted.

## 2.2 *Prototype Tests*

For each batch, the following tests shall be conducted in sequence on five timepieces or the appropriate components. If the batch is less than 20, tests need be conducted on only one timepiece.

### 2.2.1 Type of nuclide, activity and marking

- (a) *Type of nuclide* : It should be determined by standard methods and/or by certification of the supplier of the radioisotope.
- (b) *The activity* shall be determined by methods appropriate for the radionuclide<sup>1</sup>.
- (c) *Marking* : Visual inspection.

### 2.2.2 Adhesion, release of activity and solubility

- (a) *Adhesion* : The dials shall be attached to a vibrating fixture and undergo vibration at a rate of between 20 and 30 cycles per second, and a vibration acceleration of not less than 2 g for a period of not less than one hour (g is the acceleration due to the earth's gravity).

The hands shall be bent over a cylinder of 2.5 cm diameter. If the length of the hand exceeds 1.5 cm, a larger bending radius up to the length of the hand tested shall be permitted. If for reasons of special design, bending tests are not practicable, they should be replaced by a vibration test similar to that for dials.

After this, appropriate tests should be made to check the firm adhesion of the paints on the dials and hands, i.e. by means of U-V lamps and/or measurement of the activity of the painted components in order to ensure that no significant loss of activity had occurred.

- (b) *Release of activity and solubility for Tritium-activated paints* : Hands and dials containing Tritium-activated paints which have been subject to vibration or bending tests shall be totally immersed in distilled water at  $20 \pm 2^\circ\text{C}$  for 24 hours. The water must stand at least 3 mm above the painted area. The Tritium content of the water shall not exceed 5 % of the original activity of the tested components.

### 2.2.3 Casing

The transparent cover must have a thickness equivalent to not less than  $50 \text{ mg/cm}^2$  at any point.

## 2.3 *Quality Control*

Quality control procedures shall be used to ensure that the quality of the timepiece or components compares adequately with the quality of those timepieces or components employed in the prototype tests. As a minimum, quality control shall consist of 100% visual inspection to detect cracking or flaking of paint, imperfections in the transparent cover, inadequate markings, etc.

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1. For instance : by measuring of the activity or of the luminosity, applying chemical procedures and other appropriate methods.

Special control procedures may be set out by the appropriate national authority, which shall take any measures necessary to ensure compliance with the standards. The manufacturer should make any additional tests necessary to fulfil the Standards.

### *Annex I*

#### *Selection of Radionuclides to be used in Radioluminous Timepieces*

##### 1. *General criteria for selection*

Out of the large number of naturally occurring and artificially produced radionuclides, only those which possess the following properties are technically suitable for use in radioluminous paint :

- (i) Emit radiation suitable for producing luminosity in luminous paint, but which do not cause significant deterioration of the phosphor.
- (ii) Have a lifetime long enough so that sufficient luminosity is obtained during the lifetime of the timepiece for which they are used.
- (iii) Can be readily obtained at moderate costs in the required concentration and quantity.

The selection of radionuclides which can be permitted for use in radioluminous paint for timepieces must take into account the particular requirements of radiation protection which may be summarised as follows :

The quantity of the radionuclide per timepiece required to obtain sufficient luminosity shall be so low that :

- (i) the doses which would result from external radiation of the timepiece should constitute only an insignificant fraction of the maximum permissible doses which apply to individual members of the public<sup>1</sup> ;
- (ii) the doses which would result from internal radiation if particles of the radioluminous paint were accidentally incorporated should — even under extreme conditions of intake — not constitute a significant radiation hazard.

For convenience of physical surveillance it is also desirable that the radionuclide meets the following requirements :

- (i) It can be obtained without significant contamination from other radionuclides.
- (ii) It can be bound in the luminous paint so that it does not escape in significant amounts.
- (iii) It can be easily measured in quantities which are to be kept under control (e.g. activity handled, surface contamination, concentration in air and water, body burden).

In Table 1 are listed those radionuclides which have been used so far or which have been considered for use. As the following radionuclides are not commercially available, they are not included in the table : Sm-151, Tm-171, Ac-227, U-232, U-233, Np-237, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Cm-244, Cm-246, Cf-250, Cf-252. Ra-228 has been omitted because it can be obtained only with varying contents of Ra-226.

For the assessment of doses from external radiation are listed the specific gamma-ray emission and the specific beta-ray emission of the radionuclides.

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1. This fact stresses once again the desirability of using Pm-147 and H-3 (see point 4.1.1).

Doses from internal radiation can be assessed for all the radionuclides included in Table 1 from data published by ICRP [1].

## 2. Activity required to produce adequate luminosity

The basic concept in radiation protection is that all doses be kept as low as practicable and that any unnecessary exposure be avoided. In radiation protection applicable to the use of radioluminous timepieces compliance with the basic concept of radiation protection is best achieved if the activity per timepiece is kept as low as practicable, i.e. not higher than is required to produce adequate luminosity.

The recognition of hand positions and dial divisions by luminous paint marks depends primarily on the size of the marking, the brightness of the surface<sup>1</sup> and the reading distance. The values of minimum brightness listed in Table 2 were assessed [2] under the assumption that «point marking» is applied and that the spot diameters and reading distances listed in the second and third column respectively apply to the various types of timepieces for which the values are given. It is to be noted that also normal eyesight and adaptation of the eye to darkness over a period of 30 minutes was presupposed in the assessment of the values of minimum brightness. Investigations of the activity required per gram of phosphor to produce a brightness in the range of 1  $\mu\text{L}$  to 100  $\mu\text{L}$  have been published for H-3, Sr-90/Y-90, Pm-147, Tl-204 and Ra-226 by various authors [3, 4, 5] and by luminous paint manufacturers [6, 11]<sup>2</sup>. On the basis of these values the activity ratio

$$R_A = \frac{\text{activity of beta-emitter / gram phosphor}}{\text{activity of Ra-226 / gram phosphor}}$$

which corresponds to luminous densities of 1 to about 10  $\mu\text{L}$  was assessed [2].

The values of the activity per gram phosphor (ZnS/Ag) required to produce an initial brightness of 1  $\mu\text{L}$  (two months after manufacture)  $A(0)$  and the corresponding activity ratios  $R_A(0)$  listed in columns 2 and 3 of Table 3 do not make allowance for losses of brightness due to radioactive decay, release of activity and deterioration of the phosphor during the lifetime of the timepiece, and due to the absorption of radiation and visible light in the painted spot. These losses were taken into account via time dependant loss factors  $\tau$  (radioactive decay) and  $\eta$  (deterioration of the phosphor and release of activity from the paint), and the time independant loss factor  $\delta$  (loss in the applied condition as compared to the brightness of the activated natural phosphor in powder form).

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1. Luminosity is measured in units of brightness. Brightness is measured by the flux emitted per unit emissive area as projected on a plane normal to the line of sight. The unit of brightness is that of a perfectly diffusing surface giving out one lumen per square centimeter of projected surface and is called the lambert (L).

2. It is to be noted that the reported values are not in all cases suitable for comparison. Different phosphors were used and the method of paint preparation and of measurement of luminosity were not standardized. For that reason the calculations of the activities required to produce a luminosity of 1  $\mu\text{L}$  had to be based on the best possible interpolation from measured data. Therefore the ratios listed should not be taken as absolute values, they are intended for guidance only in the assessment of the order of magnitude of exposure levels from external and internal radiation and would require correction as more detailed information becomes available.

The values  $\tau(10)$ ,  $\tau_i(10)$ ,  $\lambda(10)$ ,  $A(10)$  and  $R_A(10)$  given in Table 3 were assessed<sup>[2]</sup> for timepieces with an expected lifetime of 10 years. The influence of colour additives, which reduce the luminous density to as much as 30% of the uncoloured phosphor, was left out of account, because such colourings conflict with the basic concept of radiation protection. On the basis of the  $A(10)$  values given in Table 3 and values quoted by the dial painting industry for the quantities of activated phosphor required per timepiece and the minimum luminous densities needed for the various types of ordinary timepieces (see Table 2) an assessment was made of the total activity required per timepiece (see Table 4). These values are found to be some 3-4 times higher for travel alarm clocks and large alarm clocks than for other timepieces.

It should be noted that the values of the total activity of timepieces listed in Table 4 are minimum values. Allowance should be made for higher activities in order to compensate for variation of eyesight, to reduce the time required for adaption of the eye for reading and to compensate for quality variation of the luminous paint. For that reason four times the minimum total activity values which apply to small watches and twice the minimum total activity values which apply to alarm clocks (in these cases variation of eyesight can be compensated by reduction of the reading distance) may be used for guidance in limiting the total activity of ordinary timepieces. For special timepieces ten times the minimum total activity values which apply to small watches may be considered a reasonable upper limit of the total activity per timepiece. It should be possible that with these activity levels the luminosity can be obtained which is required to permit reading of the time under all the special conditions for which special timepieces are designed.

### 3. Hazards of exposure from external and internal radiation

Due to more than 60 years of experience with Radium, the potential risks of radiation exposure from this element and its daughter products are well understood. Timepieces with luminous paint activated with Radium have been in use over the past 50 years and there exists a considerable volume of literature which gives estimates of radiation doses to the public resulting from the use of Radium-activated luminous timepieces. It would seem useful to make use of these data in arriving at comparative estimates of the radiological hazards to the public resulting from the use of luminous timepieces containing the other radionuclides which are listed in Table 1.

#### 3.1 External exposure

The only data at present available on the doses received by various organs as a result of external exposure from timepieces relate to timepieces containing Ra-226 <sup>[12, 22]</sup>. Experimental investigations have shown that the radiation doses are closely bound up with the habits of the user and the design of the timepiece. Values for specific beta- and gamma-ionization (see Table 1) and the relative activity values  $R_A(10)$  (see Table 3) are therefore used here in assessing the external exposure to be expected from the use of the radionuclide X under consideration, relative to the exposure which must

be expected if Ra-226 is used<sup>1</sup>. In calculating the  $K_{\beta}$  values it was assumed that beta-radiation is absorbed by 50 mg/cm<sup>2</sup> (minimum thickness of the transparent cover). Contributions from bremsstrahlung and absorption in air were ignored. The decay of the radionuclide over the lifetime of the timepiece is taken into account by introducing the integral mean value of the specific beta- and gamma-ray emission over that period. The relative dose values shown in Table 5 correspond to the relationship

$$R_{\beta,\gamma}(10) = \frac{R_A(10) \times K_{\beta,\gamma} \times \frac{1 - e^{-0.693t/T_p}}{0.693t/T_p}}{[K_{\beta,\gamma}] \text{ Ra-226}} \times X$$

$R_A(10)$  — Ratio of the specific activity of the given radionuclide (Ci/g of phosphor) required to produce a luminosity of 1  $\mu$ L after a period of 10 years to the specific activity of Radium-226 required to produce a luminosity of 1  $\mu$ L after a period of 10 years.

$R_{\beta}(10)$ ,  $R_{\gamma}(10)$  — Ratio of the external beta or gamma radiation exposure received over a period of 10 years from a specific activity  $R_A(10)$  of the given radionuclide to the external beta or gamma radiation exposure received over a period of 10 years from unit specific activity of Radium-226.

The values of  $R_{\beta}$  and  $R_{\gamma}$  can only serve as a rough guide in the assessment of surface doses. The assessment of the doses received by specific organs would require more elaborate calculations whereby in particular the exposure geometry and the absorption of the radiation in tissue would need detailed consideration. It can be seen from Table 5 that exposure of the user of a timepiece from beta radiation and gamma radiation can be reduced to zero level if H-3, C-14, Ni-63 or Pm-147 are used. In the case of Tc-99 if the thickness of the absorbent medium is assumed to be 75 mg/cm<sup>2</sup>, the value of  $R_{\beta}$  would be barely distinguishable from zero. However, if Cl-36, Kr-85, Sr-90 or Tl-204 were used, then the exposure from beta radiation would be significantly (by factors between 20 and 70) higher than that which would result from use of Ra-226. A beta dose still higher by a factor of 5 would result from the use of Pb-210. In case of Th-230 and Am-241 there would not exist exposure from beta radiation and the doses from gamma radiation could be considerably reduced.

### 3.2 Internal exposure

Table 6 shows dose equivalent values (rem) per unit activity ( $\mu$ Ci) taken up by adults (standard man). The values were calculated for various modes of uptake of soluble and insoluble materials on the basis of the values published by ICRP (1) for metabolic behaviour, effective energy and organ weights. For effective energies whose effective half-lives are long in com-

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1. There exist some uncertainties with respect to the  $R_{\beta}$  value of Ra-226. If it is assumed that Bi-210 (RaE), daughter product of Pb-210 (RaD) ( $T_p = 19.4y$ ) is not present, then the calculated value is  $K_{\beta} = 370 \frac{\text{rad cm}^2}{\text{h mCi}}$ . In calculating the dose ratio, the value  $K_{\beta} = 400 \frac{\text{rad cm}^2}{\text{h mCi}}$  was used for Ra-226.

parison with the expected exposure times (Sr-90, Ra-226, Th-230 and Am-241) the dose which would be received after a period of 50 years is not taken into account. The doses for uptake of soluble radionuclide through wounds ( $D_{WS}$ ) and for the inhalation of the insoluble radionuclide ( $D_{II}$ ) correspond to Fairbairn's [23] data. The values for the ingestion of soluble radionuclides ( $D_{OS}$ ) and the inhalation of soluble radionuclides ( $D_{IS}$ ) were calculated on the basis of the following relationships :

$$D_{OS} = f_1 \times D_{WS}$$

$$D_{IS} = \frac{f_a}{f_w} \times D_{OS}$$

where  $f_1$  = fraction of the radionuclide which passes from the gastrointestinal tract to the blood

$f_a$  = fraction of the radionuclide which reaches the reference organ following inhalation

$f_w$  = fraction of the radionuclide which reaches the reference organ following ingestion.

The dose values for ingestion of insoluble radionuclides ( $D_{OI}$ ) were obtained from the formula

$$D_{OI} = \frac{19.2 \times \Sigma E \text{ (RBE)}}{m}$$

where  $\Sigma E \text{ (RBE)}$  = effective energy for the reference organ area of the gastrointestinal tract, and

$m$  = mass of the reference area of the gastrointestinal tract (lower large intestine).

The doses given in Table 6 together with the activity values  $R_A$  (Table 3) provide a basis for the assessment of the relative internal radiation exposure, according to the mode of uptake for the radionuclides under consideration.

The relative doses

$$R_{OI} = \frac{(R_A \times D_{OI}) \times}{(D_{OS}) \text{ Ra-226}} ; \text{ (ingestion in insoluble form)}$$

$$R_{II} = \frac{(R_A \times D_{II}) \times}{(D_{IS}) \text{ Ra-226}} ; \text{ (inhalation in insoluble form)}$$

$$R_{OS} = \frac{(R_A \times D_{OS}) \times}{(D_{OS}) \text{ Ra-226}} ; \text{ (ingestion in soluble form)}$$

$$R_{IS} = \frac{(R_A \times D_{IS}) \times}{(D_{IS}) \text{ Ra-226}} ; \text{ (inhalation in soluble form)}$$

$$R_{WS} = \frac{(R_A \times D_{WS}) \times}{(D_{WS}) \text{ Ra-226}} ; \text{ (uptake through wounds in soluble form)}$$



show the factor by which the dose corresponding to absorption of luminous paint from a timepiece containing radionuclide X differs from that due to the absorption of a similar amount of luminous paint containing a Ra-226 activated phosphor.

All these dose ratios are based on the soluble form of Ra-226, because it must be assumed that Ra-226 activated paints produced so far cannot be considered completely insoluble [24].

The values calculated from activity ratios which apply to timepieces with a lifetime of 10 years are given in Table 7. The absorption of insoluble materials through wounds has not been considered because it can be assumed that most of the material is removed from the wound during treatment and therefore does not reach the critical organ.

It can be deduced from Table 7 that

- (i) The dose from internal radiation in the case of ingestion would be less for each of the available radionuclides than for soluble Ra-226, provided that the radionuclide is used in the insoluble form. In the case of Ra-226 the dose from internal radiation would result only in about 1/500 of the dose to be expected from use of the soluble product.
- (ii) With the available radionuclides the dose from internal radiation caused by inhalation of insoluble radionuclides, except Pb-210, would be less than or comparable to the dose to be expected from the use of Ra-226 in its soluble form.
- (iii) Except in the case of Sr-90, the dose from internal radiation after ingestion of the soluble form of the available radionuclides would be less than the dose to be expected if soluble Ra-226 were used.
- (iv) When the available radionuclides are used, the dose from internal radiation resulting from inhalation of the soluble radionuclides (with the exception of Sr-90, Pb-210, Th-230 and Am-241) would be less than the dose to be expected from the use of soluble Ra-226.
- (v) With the exception of Sr-90, Pm-147, Pb-210, Th-230 and Am-241, the dose from internal radiation resulting from uptake of radionuclides in soluble form through wounds would be less than the dose to be expected from the use of soluble Ra-226.
- (vi) Ra-226 could only be considered less dangerous than other radionuclides if it could be kept in insoluble form in radioluminous paints.

#### 4. *Conclusions*

Primary objective of the selection of radionuclides should be to prevent foreseeable risks, such as exposure from external radiation. External exposure from timepieces can be kept at zero level if H-3, C-14, Ni-63 or Pm-147 are used and at insignificant levels if Th-230 or Am-241 are used. Therefore these radionuclides would be suitable for use in timepieces provided that there do not exist significant risks of internal radiation exposure in case of an accident. In case of H-3, C-14, Ni-63 and Pm-147 there do not exist such excessive high risks regardless whether soluble or insoluble material is used. However, in the case of Th-230 and Am-241 such risks would exist if the material were to be inhaled or incorporated through wounds in its soluble form. Due to the relatively high costs and/or due to problems in the production of the required quantities and concentrations of C-14 and Ni-63 limited practical experience of their use for radioluminous paint is available.

Therefore a decision whether the use of these radionuclides in radioluminous paint for timepieces can be recommended must be postponed until more experience is obtained. This leaves as radionuclides which can be recommended for use in timepieces

### H-3 and Pm-147.

As regards Ra-226 it is appreciated that the exposure risks are well understood and can well be kept under control. The exposure levels from beta- and gamma-radiation and radon leakage do not present a significant hazard if the total activity is kept at levels which are subject to a detailed study in Annex II of this manual and if adequate measures are taken which prevent unduly high radon leakage. Taking into account the worldwide use of Radium for radioluminous paint in timepieces during a period of about 50 years which has not led to a significant contribution to the exposure of human beings [25], further use of Radium-226 can be permitted.

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TABLE 1  
 TECHNICAL DATA OF RADIONUCLIDES CONSIDERED  
 FOR USE IN RADIOLUMINOUS TIMEPIECES

| Radionuclide   | Half Life<br>(years) | Type of<br>Radiation<br>emitted | $K_{\gamma}$  | $K_{\beta}$  |
|----------------|----------------------|---------------------------------|---|--|
|                |                      |                                 | Specific<br>gamma-ray<br>emission<br>$\frac{R}{h} \frac{cm^2}{mCi}$ | Specific<br>beta-ray<br>emission (1)<br>$\frac{rad}{h} \frac{cm^2}{mCi}$ |
| H-3            | 12.46                | $\beta$                         | —   | 0  |
| C-14           | $5.57 \times 10^3$   | $\beta$                         | —   | 0  |
| Cl-36          | $3.1 \times 10^5$    | $\beta, K$                      | —   | 150  |
| Ni-63          | 125                  | $\beta$                         | —   | 0  |
| Kr-85/Rb-85m   | 10.27                | $\beta, \gamma$                 | 0.021   | 140  |
| Sr-90/Y-90     | 28                   | $\beta, \gamma$                 | 0.0017  | 370  |
| Tc-99          | $2.1 \times 10^5$    | $\beta$                         | —   | 18   |
| Pm-147/Sm-147  | 2.50                 | $\beta$                         | —   | <2.8   |
| Tl-204         | 3.57                 | $\beta, K$                      | 0.007   | 160  |
| Pb-210         | 19.4                 | $\alpha, \beta, \gamma$         | 0.014   | 205  |
| Ra-226         | $1.62 \times 10^3$   | $\alpha, \beta, \gamma$         | 9.3   | 580  |
| Th-230         | $8.0 \times 10^4$    | $\alpha, \gamma$                | 0.0033  | 0  |
| Am-241/Np-237m | 470                  | $\alpha, \gamma$                | 0.14  | 0  |

1. 50 mg/cm<sup>2</sup> absorbent medium.

TABLE 2  
 MINIMUM BRIGHTNESS REQUIRED FOR VARIOUS TYPES  
 OF ORDINARY TIMEPIECES

| Types of timepiece   | spot<br>diameter<br>(mm) | reading<br>distance<br>(cm) | minimum<br>brightness<br>( $\mu L$ ) |
|----------------------|--------------------------|-----------------------------|--------------------------------------|
| Ladies' wristwatches | 0.4 — 0.8                | 25                          | 0.38 — 1.4                           |
| Mens' wristwatches   | 0.6 — 1.0                | 25                          | 0.24 — 0.65                          |
| Pocket watches       | 0.8 — 1.0                | 25                          | 0.24 — 0.38                          |
| Travel alarm clocks  | 1.0 — 1.5                | 50                          | 0.44 — 0.97                          |
| Large alarm clocks   | 1.5 — 3.0                | 50                          | 0.11 — 0.44                          |

TABLE 3

Activity per gram phosphor (ZnS/Ag) [Ci/g] required for producing a luminosity of  $1 \mu\text{L}$

(i) shortly (2 months) after production of the radioluminous paint :  $A(0)$

(ii) 10 years after production of the radioluminous paint :  $A(10) = A(0)/\sigma$  and the corresponding activity ratios  $R_A(0)$ ,  $R_A(10)$

| Radionuclide | A(0)<br>(Ci/g)       | $R_A(0)$          | Loss Factor |            |          | Total loss<br>$\sigma(10)$ | A(10)<br>(Ci/g)      | $R_A(10)$          |
|--------------|----------------------|-------------------|-------------|------------|----------|----------------------------|----------------------|--------------------|
|              |                      |                   | $\tau(10)$  | $\eta(10)$ | $\delta$ |                            |                      |                    |
| H-3          | $4.6 \times 10^{-3}$ | $5.3 \times 10^3$ | 0.56        | 0.27       | 0.18     | 0.027                      | $1.7 \times 10^{-1}$ | $2.00 \times 10^4$ |
| C-14         | $1.9 \times 10^{-4}$ | $2.2 \times 10^2$ | 1.00        | 0.21       | 0.37     | 0.078                      | $2.4 \times 10^{-3}$ | $2.8 \times 10^2$  |
| Cl-36        | $4.3 \times 10^{-5}$ | $5.0 \times 10^1$ | 1.00        | 0.15       | 0.50     | 0.075                      | $5.7 \times 10^{-4}$ | $6.5 \times 10$    |
| Ni-63        | $5.7 \times 10^{-4}$ | $6.7 \times 10^2$ | 0.94        | 0.25       | 0.30     | 0.070                      | $8.1 \times 10^{-3}$ | $9.3 \times 10^2$  |
| Kr-85        | $4.7 \times 10^{-5}$ | $5.5 \times 10^1$ | 0.52        | 0.16       | 0.50     | 0.042                      | $1.1 \times 10^{-3}$ | $1.3 \times 10^2$  |
| Sr-90        | $1.9 \times 10^{-5}$ | $2.2 \times 10^1$ | 0.77        | 0.11       | 0.65     | 0.055                      | $3.4 \times 10^{-4}$ | $3.9 \times 10$    |
| Tc-99        | $1.0 \times 10^{-4}$ | $1.2 \times 10^2$ | 1.00        | 0.16       | 0.42     | 0.067                      | $1.5 \times 10^{-3}$ | $1.7 \times 10^2$  |
| Pm-147       | $1.5 \times 10^{-4}$ | $1.7 \times 10^2$ | 0.068       | 0.20       | 0.39     | 0.0053                     | $2.8 \times 10^{-2}$ | $3.2 \times 10^3$  |
| Tl-204       | $4.3 \times 10^{-5}$ | $5.0 \times 10^1$ | 0.17        | 0.16       | 0.50     | 0.013                      | $3.2 \times 10^{-3}$ | $3.7 \times 10^2$  |
| Pb-210       | $3.9 \times 10^{-6}$ | 4.5               | 0.71        | 0.16       | 0.35     | 0.040                      | $9.8 \times 10^{-5}$ | $1.1 \times 10$    |
| Ra-226       | $8.5 \times 10^{-7}$ | 1.0               | 1.00        | 0.26       | 0.38     | 0.098                      | $8.7 \times 10^{-6}$ | 1.0                |
| Th-230       | $4.3 \times 10^{-6}$ | 5.1               | 1.00        | 0.30       | 0.35     | 0.105                      | $4.1 \times 10^{-5}$ | 4.7                |
| Am-241       | $3.7 \times 10^{-6}$ | 4.4               | 1.00        | 0.30       | 0.35     | 0.105                      | $3.5 \times 10^{-5}$ | 4.0                |

TABLE 4  
MINIMUM TOTAL ACTIVITY (mCi) OF VARIOUS TYPES OF TIMEPIECES

| Type of time-piece              | Ladies' Wrist-watches | Mens' Wrist-watches  | Pocket Watches       | Travel Alarm Clocks  | Large Alarm Clocks   |
|---------------------------------|-----------------------|----------------------|----------------------|----------------------|----------------------|
| Spot diameter (mm)              | 0.6                   | 0.8                  | 0.9                  | 1.3                  | 2.3                  |
| Total quantity of phosphor (mg) | 4                     | 7                    | 9                    | 18                   | 50                   |
| Reading distance (cm)           | 25                    | 25                   | 25                   | 50                   | 50                   |
| Minimum brightness ( $\mu$ L)   | 0.65                  | 0.38                 | 0.30                 | 0.57                 | 0.20                 |
| Radio-nuclide                   | total activity (mCi)  |                      |                      |                      |                      |
| H-3                             | 4.4x10 <sup>-1</sup>  | 4.5x10 <sup>-1</sup> | 4.6x10 <sup>-1</sup> | 1.7                  | 1.7                  |
| C-14                            | 6.2x10 <sup>-3</sup>  | 6.4x10 <sup>-3</sup> | 6.5x10 <sup>-3</sup> | 2.5x10 <sup>-2</sup> | 2.4x10 <sup>-2</sup> |
| Cl-36                           | 1.5x10 <sup>-5</sup>  | 1.5x10 <sup>-3</sup> | 1.5x10 <sup>-3</sup> | 5.9x10 <sup>-3</sup> | 5.7x10 <sup>-3</sup> |
| Ni-63                           | 2.1x10 <sup>-2</sup>  | 2.2x10 <sup>-2</sup> | 2.2x10 <sup>-2</sup> | 8.3x10 <sup>-2</sup> | 8.1x10 <sup>-2</sup> |
| Kr-85                           | 2.9x10 <sup>-3</sup>  | 2.9x10 <sup>-3</sup> | 3.0x10 <sup>-3</sup> | 1.1x10 <sup>-2</sup> | 1.1x10 <sup>-2</sup> |
| Sr-90                           | 8.8x10 <sup>-4</sup>  | 9.1x10 <sup>-4</sup> | 9.2x10 <sup>-4</sup> | 3.5x10 <sup>-3</sup> | 3.4x10 <sup>-3</sup> |
| Tc-99                           | 3.9x10 <sup>-3</sup>  | 4.0x10 <sup>-3</sup> | 4.0x10 <sup>-3</sup> | 1.5x10 <sup>-2</sup> | 1.5x10 <sup>-2</sup> |
| Pm-147                          | 7.3x10 <sup>-2</sup>  | 7.5x10 <sup>-2</sup> | 7.6x10 <sup>-2</sup> | 2.9x10 <sup>-1</sup> | 2.8x10 <sup>-1</sup> |
| Tl-204                          | 8.3x10 <sup>-3</sup>  | 8.5x10 <sup>-3</sup> | 8.6x10 <sup>-3</sup> | 3.3x10 <sup>-2</sup> | 3.2x10 <sup>-2</sup> |
| Pb-210                          | 2.5x10 <sup>-4</sup>  | 2.6x10 <sup>-4</sup> | 2.6x10 <sup>-4</sup> | 1.0x10 <sup>-3</sup> | 9.8x10 <sup>-4</sup> |
| Ra-226                          | 2.3x10 <sup>-5</sup>  | 2.3x10 <sup>-5</sup> | 2.3x10 <sup>-5</sup> | 8.9x10 <sup>-5</sup> | 8.7x10 <sup>-5</sup> |
| Th-230                          | 1.1x10 <sup>-4</sup>  | 1.1x10 <sup>-4</sup> | 1.1x10 <sup>-4</sup> | 4.2x10 <sup>-4</sup> | 4.1x10 <sup>-4</sup> |
| Am-241                          | 9.1x10 <sup>-5</sup>  | 9.3x10 <sup>-5</sup> | 9.4x10 <sup>-5</sup> | 3.6x10 <sup>-4</sup> | 3.5x10 <sup>-4</sup> |

TABLE 5  
EXTERNAL EXPOSURE FROM BETA-RADIATION  
AND GAMMA-RADIATION

(see text for definition of  $R_\beta$  and  $R_\gamma$ )

| Radionuclide | $R_\beta$ (10) | $R_\gamma$ (10)      |
|--------------|----------------|----------------------|
| H-3          | 0              | 0                    |
| C-14         | 0              | 0                    |
| Cl-36        | 24             | 0                    |
| Ni-63        | 0              | 0                    |
| Kr-85        | 33             | 2.1x10 <sup>-1</sup> |
| Sr-90        | 32             | 6.3x10 <sup>-3</sup> |
| Tc-99        | 7.7            | 0                    |
| Pm-147       | 0              | 0                    |
| Tl-204       | 66             | 1.2x10 <sup>-1</sup> |
| Pb-210       | 4.8            | 1.4x10 <sup>-2</sup> |
| Ra-226       | 1.0            | 1.0                  |
| Th-230       | 0              | 1.7x10 <sup>-3</sup> |
| Am-241       | 0              | 6.0x10 <sup>-2</sup> |

TABLE 6  
DOSE EQUIVALENT (REM) PER UNIT ACTIVITY ( $\mu$  Ci) TAKEN  
INTO THE BODY FOR ADULTS (STANDARD MAN)

| Radio-nuclide | D <sub>WS</sub><br>(rem/ $\mu$ Ci) | Critical <sup>(1)</sup><br>organ | f <sub>l</sub>   | D <sub>OS</sub><br>(rem/ $\mu$ Ci) | $\frac{f_a}{f_w}$   | D <sub>IS</sub><br>(rem/ $\mu$ Ci) | D <sub>01</sub> <sup>(2)</sup><br>(rem/ $\mu$ Ci) | D <sub>II</sub> <sup>(3)</sup><br>(rem/ $\mu$ Ci) |
|---------------|------------------------------------|----------------------------------|------------------|------------------------------------|---------------------|------------------------------------|---|---|
| H-3           | 2.4x10 <sup>-4</sup>               | body tissue                      | 1.0              | 2.4x10 <sup>-4</sup>               | 1.0                 | 2.4x10 <sup>-4</sup>               | 1.3x10 <sup>-3</sup>                              | 1.0x10 <sup>-2</sup>                              |
| C-14          | 2.8x10 <sup>-3</sup>               | bone                             | 1.0              | 2.8x10 <sup>-3</sup>               | 0.80                | 2.2x10 <sup>-3</sup>               | 6.9x10 <sup>-3</sup>                              | 6.1x10 <sup>-2</sup>                              |
| Cl-36         | 7.0x10 <sup>-3</sup>               | whole body                       | 1.0              | 7.0x10 <sup>-3</sup>               | 0.75                | 5.2x10 <sup>-3</sup>               | 3.3x10 <sup>-2</sup>                              | 2.9x10 <sup>-1</sup>                              |
| Ni-63         | 2.4x10 <sup>-1</sup>               | bone                             | 0.3              | 7.2x10 <sup>-2</sup>               | 1.33                | 9.6x10 <sup>-2</sup>               | 2.7x10 <sup>-3</sup>                              | 2.4x10 <sup>-2</sup>                              |
| Kr-85         | —                                  | —                                | —                | —                                  | —                   | —                                  | 2.8x10 <sup>-2</sup>                              | 2.6x10 <sup>-1</sup>                              |
| Sr-90         | 9.0x10                             | bone                             | 0.3              | 2.7x10                             | 1.33                | 3.6x10                             | 1.4x10 <sup>-1</sup>                              | 1.2   |
| Tc-99         | 6.0x10 <sup>-3</sup>               | kidneys                          | 0.5              | 3.0x10 <sup>-3</sup>               | 1.0                 | 3.0x10 <sup>-3</sup>               | 1.2x10 <sup>-2</sup>                              | 1.1x10 <sup>-1</sup>                              |
| Pm-147        | 7.0x10 <sup>-1</sup>               | bone                             | 10 <sup>-4</sup> | 7.0x10 <sup>-5</sup>               | 2.6x10 <sup>3</sup> | 1.8x10 <sup>-1</sup>               | 8.8x10 <sup>-3</sup>                              | 7.0x10 <sup>-2</sup>                              |
| Tl-204        | 3.0x10 <sup>-2</sup>               | kidneys                          | 0.45             | 1.4x10 <sup>-2</sup>               | 1.04                | 1.5x10 <sup>-2</sup>               | 3.2x10 <sup>-2</sup>                              | 2.6x10 <sup>-1</sup>                              |
| Pb-210        | 2.0x10 <sup>2</sup>                | bone                             | 0.08             | 1.6x10                             | 4.0                 | 6.4x10                             | 1.2x10 <sup>-1</sup>                              | 2.8x10  |
| Ra-226        | 1.0x10 <sup>3</sup>                | bone                             | 0.3              | 3x10 <sup>2</sup>                  | 0.75                | 2.3x10 <sup>2</sup>                | 5.9x10 <sup>-1</sup>                              | 1.3x10 <sup>2</sup>                               |
| Th-230        | 1.1x10 <sup>5</sup>                | bone                             | 10 <sup>-4</sup> | 1.1x10                             | 2.6x10 <sup>3</sup> | 2.9x10 <sup>4</sup>                | 6.0x10 <sup>-2</sup>                              | 5.4x10  |
| Am-241        | 9.0x10 <sup>3</sup>                | bone                             | 10 <sup>-4</sup> | 9.0x10 <sup>-1</sup>               | 2.5x10 <sup>3</sup> | 2.3x10 <sup>3</sup>                | 7.7x10 <sup>-2</sup>                              | 6.5x10  |

1. These values relate to D<sub>WS</sub>, D<sub>OS</sub> and D<sub>IS</sub>
2. The critical organ is the lower large intestine
3. Critical organ is the lung

TABLE 7  
RELATIVE VALUES OF THE DOSE FROM INTERNAL RADIATION  
(Single intake) (See text for definition of quotients)

| Radio-nuclide | R <sub>OI</sub> (10)  | R <sub>II</sub> (10) | R <sub>OS</sub> (10) | R <sub>IS</sub> (10) | R <sub>WS</sub> (10) |
|---------------|-----------------------|----------------------|----------------------|----------------------|----------------------|
| H-3           | 8.5x10 <sup>-2</sup>  | 8.5x10 <sup>-1</sup> | 1.6x10 <sup>-2</sup> | 2.0x10 <sup>-2</sup> | 4.7x10 <sup>-3</sup> |
| C-14          | 6.4x10 <sup>-3</sup>  | 7.4x10 <sup>-2</sup> | 2.6x10 <sup>-3</sup> | 2.7x10 <sup>-3</sup> | 7.8x10 <sup>-4</sup> |
| Cl-36         | 7.2x10 <sup>-3</sup>  | 8.3x10 <sup>-2</sup> | 1.5x10 <sup>-3</sup> | 1.5x10 <sup>-3</sup> | 4.6x10 <sup>-4</sup> |
| Ni-63         | 8.4x10 <sup>-3</sup>  | 9.7x10 <sup>-2</sup> | 2.2x10 <sup>-1</sup> | 3.9x10 <sup>-1</sup> | 2.2x10 <sup>-1</sup> |
| Kr-85         | 1.2x10 <sup>-2</sup>  | 1.5x10 <sup>-1</sup> | —                    | —                    | —                    |
| Sr-90         | 1.8x10 <sup>-2</sup>  | 2.0x10 <sup>-1</sup> | 3.5                  | 1.5x10               | 3.5                  |
| Tc-99         | 6.8x10 <sup>-3</sup>  | 8.1x10 <sup>-2</sup> | 1.7x10 <sup>-3</sup> | 2.2x10 <sup>-3</sup> | 1.0x10 <sup>-3</sup> |
| Pm-147        | 9.3x10 <sup>-2</sup>  | 9.7x10 <sup>-1</sup> | 7.5x10 <sup>-4</sup> | 2.5                  | 2.2                  |
| Tl-204        | 3.94x10 <sup>-2</sup> | 4.2x10 <sup>-1</sup> | 1.7x10 <sup>-2</sup> | 2.4x10 <sup>-2</sup> | 1.1x10 <sup>-2</sup> |
| Pb-210        | 4.4x10 <sup>-3</sup>  | 1.3                  | 5.9x10 <sup>-1</sup> | 3.1                  | 2.2                  |
| Ra-226        | 2.0x10 <sup>-3</sup>  | 5.7x10 <sup>-1</sup> | 1.0                  | 1.0                  | 1.0                  |
| Th-230        | 9.4x10 <sup>-4</sup>  | 1.1                  | 1.7x10 <sup>-1</sup> | 5.9x10 <sup>2</sup>  | 5.2x10 <sup>2</sup>  |
| Am-241        | 1.0x10 <sup>-4</sup>  | 1.1                  | 1.2x10 <sup>-2</sup> | 4.0x10               | 3.6x10               |

### *Annex II*

#### *Radiation Protection Considerations and Determination of Maximum Permissible Activities of Radionuclides recommended for Radioluminous Timepieces*

#### I. *Potential hazards resulting from the use of radioluminous timepieces*

1. Radioluminous timepieces are sources of radioactivity which may expose individual members of the population and, to some extent, the population as a whole, to external radiation on the one hand in normal conditions of use, and to the possible risk of internal contamination on the other, when protection is no longer ensured against an escape of radioactive material or contact with such material. Permanent external irradiation due to  $\beta$  and  $\gamma$  rays is produced by timepieces bearing radioluminous paint activated by Radium. If the watch glass is removed, external irradiation is also produced by  $\beta$  rays in the case of Promethium-147.

2. The activity present in a timepiece with a radioluminous dial must be as low as practicable and, in any event, lower than the amount of radionuclide which would cause the critical organ to receive the maximum permissible dose in case of accidental contamination in the most unfavourable conditions. Under normal conditions of use, the dose resulting from the presence of radionuclides in timepieces should only constitute a minor fraction of the maximum permissible dose for exposure of individual members of the population. This reduces the radioisotopes eligible for use in the manufacture of luminous timepieces — both from the health and from the current technical standpoint — to a very small number, namely Tritium, Promethium-147 and Radium-226. (See Annex I).

3. Health considerations must take into account the doses actually received from the use of timepieces, the contribution of the doses received to the genetically significant dose of the population and the doses which could



possibly be received by the individual user in case of an accident. Apart from the activity in a timepiece, the doses actually received will largely depend upon the period during which the timepiece is used.

4. For the assessment of the genetically significant dose, the number of users becomes a very important factor. For that reason, when considering the health standpoint, separate consideration will be required for ordinary timepieces, which are used continuously by large groups of the population, and for special timepieces which are normally used for limited periods by a limited number of people. However, it is anticipated that the contribution to the genetically significant dose of the population of less than 100 mremms per generation (see also Section II of this Annex), resulting from the use of ordinary radium-activated timepieces which meet the Standards set out in this manual, may seem acceptable by all national authorities.

5. As concerns special timepieces, the importance of the contribution to the genetically significant dose of the population cannot be assessed without a detailed study of the particular situation in the country concerned. Therefore, though exposure rates of external radiation from special timepieces can be assessed, it cannot be evaluated whether these exposure rates would not result in significant contributions to the genetically significant dose without introduction of some control measures. For this reason, such timepieces must, according to these Standards, be marked in order to permit to the appropriate national authority to take measures for the introduction of a control system if so deemed necessary.

6. In Section II the exposure levels from external radiation are assessed, taking into account the contributions from gamma radiation, beta radiation and bremsstrahlung which arise from the maximum activities recommended for ordinary and special timepieces.

7. In Section III the maximum exposure levels which could possibly arise from the intake of the maximum permissible activities recommended for ordinary and special timepieces are assessed. Internal radiation can be caused by inhalation or ingestion of radioactive fragments from dials which are unprotected. For the purposes of strict and efficient protection, one must assume that there is a possibility that the wearer of a luminous watch dial may inhale or ingest some of the radioactive matter from the dial at least once during a lifetime.

In order to reduce the internal irradiation hazards, the following points should be considered :

- (a) Radioactive dials and hands should be made in such a way that no undue loss of radioactivity occurs from the timepiece under foreseeable conditions. For this purpose, appropriate tests should be carried out.
- (b) Special radioluminous timepieces which are out of order and are no longer considered serviceable should be kept under control or disposed of, so that risks of contamination are most unlikely.
- (c) Radioluminous timepieces should not be given to children as toys.

8. With regard to special timepieces, the values recommended in this manual may lead, in some cases, to radiation doses exceeding those set out by the ICRP for individual members of the population, if the use of such timepieces is not strictly limited to the purposes for which they are designed.

II. *Maximum permissible activities derived from exposure limits of external radiation*

A. *Radium-226*

In practice, a risk of external radiation from radioisotopes considered permissible for producing luminescence in timepieces arises only in the case of Radium-226. The gonads and the blood-forming organs may be exposed to gamma radiation emitted by the radioactive daughter products of Radium-226. The skin of the face and the lens of the eyes could be mainly exposed to beta radiation from these daughter products. Finally, the skin of the wrist could be exposed to gamma radiation passing from the dial through the back of the watch.

Radium-226, which has been used for about 50 years for producing luminescence in zinc sulphide crystals, acts essentially by its alpha emission and by that of the daughter products with which it is more or less in equilibrium within the luminescent material. Beta emission, issuing from the daughter products, plays only a minor part in producing luminescence, but as it is not completely stopped by the glass protecting the dial, it produces external radiation the intensity of which, after passing through an average 50 mg/cm<sup>2</sup> glass, can be determined by a coefficient of specific ionization equal to 320 rads cm<sup>2</sup>/h mCi approximately<sup>1</sup>.

The gamma radiation of Radium-226 which is emitted by its daughter products is of no importance in the production of luminescence ; however, it irradiates the whole body. The exposure rates can be calculated for specific parts of the body by using the value of its specific ionization coefficient = 9 R cm<sup>2</sup>/h mCi.

In terms of these specific ionization coefficients, and of the average distance between the organ and the dial [<sup>1</sup>, <sup>2</sup>, <sup>3</sup>], it is possible to determine, for a maximum activity of 0.15  $\mu$ Ci of Radium-226, the annual radiation doses received by the critical organs : these are shown in Table 1.

TABLE 1

| Organ                             | Type of Irradiation | Annual Dose received |
|-----------------------------------|---------------------|----------------------|
| Lenses of the eyes<br>(3 mm deep) | beta                | 0.11 rad (a)         |
| Skin of the face                  | beta                | 0.4 rad (a)          |
| Blood-forming<br>tissues (trunk)  | gamma               | 0.03 rad (b)         |
| Skin of the wrist<br>(at 1.5 cm)  | gamma               | 4.8 rad (c)          |

(a) Value determined experimentally.

(b) Calculated value.

(c) Calculated value. The maximum permissible dose to the skin for individual members of the population is 7.5 rems per annum.

1. If it is assumed that all the daughter products including Po-210 (RaF) are present, in equilibrium amounts, when filtered by 50 mg/cm<sup>2</sup>, the value is about 550 rads/h and, when filtered by 100 mg/cm<sup>2</sup>, the value is about 300 rads/h. A considerable fraction of the beta emission is from Bi-210 (RaE), which will only be present in appreciable amounts many years after the preparation of the Radium, as it is a daughter of Pb-210 (RaD) which has a 20 year half-life and which is likely to be eliminated in the preparation of the Ra-226 from the mineral. If no Pb-210 is assumed to be present, the values of beta-ray emission are about 320 and 180 rads/h for the two thicknesses respectively.

It can be seen from the above table that all the critical organs receive an annual radiation dose lower than that laid down by the ICRP.

The «genetic» radiation dose resulting from exposure of the gonads to gamma radiation from luminous dials containing Radium should be determined for the population as a whole. It depends on the proportion of men and women wearing radioluminous watches, their age, child expectancy, and finally, on the annual dose received by the gonads for an activity of 0.1  $\mu$  Ci from a watch worn on the wrist<sup>1</sup>.

The ICRP has suggested a maximum population genetic dose of 5 rems from all sources, in addition to medical procedures and natural background radiation. In an illustrative apportionment of this dose, the ICRP has indicated that 0.5 rem might be allocated to the external irradiation of the population at large. This is equivalent to 17 mrems per year<sup>2</sup>.

#### B. *Promethium-147*

Pm-147 is a pure beta emitter. The maximum range of its beta particles is 46 mg/cm<sup>2</sup>. Since the Standards set out in this manual recommend a minimum thickness of 50 mg/cm<sup>2</sup> for the casing of timepieces, no exposure hazard from beta radiation should exist. The total activity per timepiece (150  $\mu$ Ci per ordinary timepiece) and the energy of the beta radiation ( $E_{\max} = 0.223$  MeV) is low enough not to produce bremsstrahlung in such amounts and energies as would require special attention. From radiation analysis of Pm-147 it is known that this radioisotope is not free from contamination by other radionuclides. In particular Eu-154, a gamma emitter, is normally present in commercially available products. However, the quantities can be kept at less than 0.004 % so that the gamma radiation can be kept at insignificant levels [4], and in any case lower than 0.1 mrem/h at 10 cm distance. In the case of pocket watches (ordinary timepieces) containing Pm-147, control measures should be applied in order to ensure that the dose level of 0.1 mrem/h at 10 cm distance is not exceeded.

#### C. *Tritium*

As Tritium is a pure beta emitter with a maximum range of beta particles of 0.6 mg/cm<sup>2</sup> the beta energy is low enough to ensure that no external radiation results from luminous watches containing Tritium.

### III. *Maximum permissible activities derived from exposure limits of internal radiation*

After introduction into the body, Tritium, Promethium-147 and Radium-226, depending on their chemical state, i.e. soluble or insoluble, follow the general metabolic channels, each lodging in one or several critical organs. Once lodged, the ionizing effect diminishes according to the effective half-life (i.e. physical half-life and biological half-life). The radiation dose received

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1. A series of studies carried out in Switzerland showed that this specific dose was equal to 70 mR per  $\mu$  Ci/year for a man and 65 mR per  $\mu$  Ci/year for a woman.

2. It emerged from a statistical study carried out in Switzerland that about 66 % of the male and 25 % of the female population wear watches with dials excited by an average activity of 0.1  $\mu$  Ci of Radium and taking into account the age and the fertility rate, this works out at a genetic dose of 100 mR per generation, i.e. 3.3. mR/year. Due to the high percentage of luminous timepieces in use in Switzerland, these values can be considered as exceptionally high and will be lower in other countries.

by the critical organ must remain below the maximum permissible doses specified in the basic Radiation Protection Norms. In estimating this dose, the chemical form of the radioactive substance — in particular as regards Radium-226 (depending on whether it is present in the form of a soluble or insoluble salt) — has to be taken into consideration. It will always be desirable that the radioisotopes should be present in as insoluble a form as possible.

According to the Report of the ICRP Committee II with regard to soluble material, Tritium settles, after ingestion or inhalation, throughout the whole body. Promethium-147 settles, through inhalation, in the bones, the amount absorbed by ingestion being negligible. Radium-226, after ingestion, lodges mainly in the bones. Radioluminous paints may be relatively insoluble and the critical organ may therefore be the gastrointestinal (GI) tract, the lung or others.

The only possible risk involved is of ingestion or inhalation of a fragment of luminescent material, or of intake through wounds, if the watch is opened or the protective glass is broken. This possible hazard is assessed in Table 2, which gives the radiation dose in rems received by the critical organs, assuming an accidental ingestion or inhalation of radioactivity equal to one-tenth of that of a watch containing the average permissible quantity <sup>(1)</sup>. The doses indicated in Table 2 are dissipated in less than twelve months in the case of Tritium, over several years for Promethium-147 (corresponding to the total disintegration of the isotope), and within fifty years for Radium-226 (following the basic ICRP concept taking fifty years in relation to the life span).

The annual doses are still below the maximum permissible dose of 0.5 rem applicable to the whole body, the gonads and the blood-forming organs. By and large, accidental absorption by an individual of a fragment of dial or hand containing the activities indicated represents a negligible hazard, and is an acceptable risk with regard to the population as a whole.

Similar considerations may be applied to ordinary clocks and special timepieces, with correspondingly greater doses because of the larger activities permitted. In all these considerations it should be borne in mind that such accidental absorption will be exceedingly rare.

TABLE 2

Dose equivalent values corresponding to various modes of intake of one-tenth of the average permissible activity recommended for ordinary watches.

| Radio-nuclide  | chemical form | mode of intake | critical organ | activity ingested ( $\mu$ Ci) | dose equivalent (rems) |
|----------------|---------------|----------------|----------------|-------------------------------|------------------------|
| H-3            | insoluble     | ingestion      | GI lung        | 500                           | $6.5 \times 10^{-1}$   |
|                |               | inhalation     |                | 500                           | 5.0                    |
|                | soluble       | ingestion      | body tissue    | 500                           | $1.05 \times 10^{-1}$  |
| inhalation     |               | body tissue    | 500            | $1.05 \times 10^{-1}$         |                        |
| through wounds |               | body tissue    | 500            | $1.05 \times 10^{-1}$         |                        |
| Pm-147         | insoluble     | ingestion      | GI lung        | 10                            | $8.8 \times 10^{-2}$   |
|                |               | inhalation     |                | 10                            | $7.0 \times 10^{-1}$   |
|                | soluble       | ingestion      | bone           | 10                            | $7.4 \times 10^{-4}$   |
| inhalation     |               | bone           | 10             | 2.0                           |                        |
| through wounds |               | bone           | 10             | 7.4                           |                        |
| Ra-226         | insoluble     | ingestion      | GI lung        | 0.01                          | $7.6 \times 10^{-3}$   |
|                |               | inhalation     |                | 0.01                          | 1.3                    |
|                | soluble       | ingestion      | bone           | 0.01                          | 3.1                    |
| inhalation     |               | bone           | 0.01           | 3.1                           |                        |
| through wounds |               | bone           | 0.01           | 10.4                          |                        |

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