**28.44 - Radioactive chemical elements and radioactive isotopes (including the fissile or fertile chemical elements and isotopes) and their compounds; mixtures and residues containing these products.**

2844.10 - Natural uranium and its compounds; alloys, dispersions (including cermets), ceramic products and mixtures containing natural uranium or natural uranium compounds

2844.20 - Uranium enriched in U 235 and its compounds; plutonium and its compounds; alloys, dispersions (including cermets), ceramic products and mixtures containing uranium enriched in U 235, plutonium or compounds of these products

2844.30 - Uranium depleted in U 235 and its compounds; thorium and its compounds; alloys, dispersions (including cermets), ceramic products and mixtures containing uranium depleted in U 235, thorium or compounds of these products

2844.40 - Radioactive elements and isotopes and compounds other than those of subheading 2844.10, 2844.20 or 2844.30; alloys, dispersions (including cermets), ceramic products and mixtures containing these elements, isotopes or compounds; radioactive residues

2844.50 - Spent (irradiated) fuel elements (cartridges) of nuclear reactors

## (I) **ISOTOPES**

The nuclei of an element, defined by its atomic number, always contain the same number of protons, but they may have different numbers of neutrons and, consequently, will be of different mass (different mass number).

Nuclides which differ only in the mass number and not in the atomic number, are called isotopes of the element. For example, there are several nuclides with the same atomic number 92 which are all called uranium, but their mass number ranges from 227 to 240; they are designated, for example, as uranium 233, uranium 235, uranium 238, etc. Analogously, hydrogen 1, hydrogen 2 or deuterium (classified in **heading 28.45**) and hydrogen 3 or tritium are isotopes of hydrogen.

The important factor in the chemical behaviour of an element is linked to the amount of the positive electric charge on the nucleus (number of protons); this determines the number of orbital electrons which actually affect the chemical properties.

Because of this, different isotopes of an element whose nuclei have the same electrical charge but different masses, will have the same chemical properties but their physical properties will vary from one isotope to another.

Chemical elements are composed either of a single nuclide (monoisotopic elements) or of a mixture of two or more isotopes in known unvarying proportions. For example, natural chlorine, in both the free and combined states, is always made up of a mixture of 75.4 % chlorine 35 and 24.6 % chlorine 37 (which gives it its atomic weight of 35.457).

When an element is composed of a mixture of isotopes, its constituent parts can be separated for example by diffusion through porous tubes, by electro-magnetic separation or by fractional electrolysis. Isotopes can also be made by bombarding natural elements with neutrons or charged particles of high kinetic energy.

For the purposes of Note 6 to this Chapter and of headings 28.44 and 28.45, the term **isotopes** covers not only isotopes in their pure state but also chemical elements whose natural isotopic composition has been artificially modified by enriching the elements in some of their isotopes (which is the same as depleting them in some others), or by converting, through a nuclear reaction, some isotopes into other, artificial isotopes. For example, chlorine of atomic weight 35.30 obtained by enriching this element to contain 85 % of chlorine 35 (and consequently by depleting it to contain 15 % of chlorine 37) is considered as an isotope.

It should be noted that elements existing in nature in the monoisotopic state, e.g., beryllium 9, fluorine 19, aluminium 27, phosphorus 31, manganese 55, etc., are not to be considered as isotopes, but are to be classified, in either the free or the combined state, according to the case, in the more specific headings relating to chemical elements or to their compounds.

Radioactive isotopes of these same elements obtained artificially (e.g. Be 10, F 18, Al 29, P 32, Mn 54) are, however, to be considered as isotopes.

Since artificial chemical elements (generally with an atomic number greater than 92, or transuranic elements) do not have a fixed isotopic composition but one which varies according to the method of obtaining the element, it is impossible in these cases to distinguish between the chemical element and its isotopes for the purposes of Note 6.

This heading covers only those isotopes which possess the phenomenon of **radioactivity** (described below); stable isotopes, on the other hand, are classified in **heading 28.45**.

## (II) RADIOACTIVITY

Certain nuclides, whose nuclei are unstable, whether in the pure state or in the form of compounds, emit complex radiations producing physical or chemical effects such as :

(1) Ionisation of gases.

(2) Fluorescence.

(3) Fogging of photographic plates.

These effects make it possible to detect these radiations and to measure their intensity by using, for example, Geiger-Müller counters, proportional counters, ionisation chambers, Wilson chambers, bubble flow counters, scintillation counters, and sensitised films or plates.

This is the phenomenon of **radioactivity**; chemical elements, isotopes, compounds and, in general, substances that display it are called **radioactive**.

## (III) RADIOACTIVE CHEMICAL ELEMENTS, RADIOACTIVE ISOTOPES AND THEIR COMPOUNDS; MIXTURES AND RESIDUES CONTAINING THESE PRODUCTS

(A) **Radioactive elements**.

Within this heading fall the radioactive chemical elements referred to in Note 6 (a) to this Chapter, namely : technetium, promethium, polonium and all elements of greater atomic number, such as astatine, radon, francium, radium, actinium, thorium, protactinium, uranium, neptunium, plutonium, americium, curium, berkelium, californium, einsteinium, fermium, mendelevium, nobelium and lawrencium.

These are elements generally composed of several isotopes which are all radioactive.

On the other hand, there are elements composed of mixtures of stable and radioactive isotopes, such as potassium, rubidium, samarium and lutetium (**heading 28.05**), which, because the radioactive isotopes have a low level of radioactivity and constitute a relatively low percentage of the mixture, can be considered as practically stable and thus do not fall in this heading.

On the other hand, the same elements (potassium, rubidium, samarium, lutetium), if enriched in their radioactive isotopes (K 40, Rb 87, Sm 147, Lu 176, respectively), are to be considered as radioactive isotopes of this heading.

(B) **Radioactive isotopes**.

To the natural radioactive isotopes potassium 40, rubidium 87, samarium 147, and lutetium 176 already mentioned, may be added uranium 235 and uranium 238, which are discussed in more detail in Section (IV) below, and certain isotopes of thallium, lead, bismuth, polonium, radium, actinium or thorium, which are often known by a name different from that of the corresponding element. This name refers rather to the element from which they were derived by radioactive conversion. Thus, bismuth 210 is called *radium E*, polonium 212 is called *thorium C'* and actinium 228 is called *mesothorium II*.

Chemical elements which are normally stable may nonetheless become radioactive either after bombardment with particles having a very high kinetic energy (protons, deuterons) issuing from a particle accelerator (cyclotron, synchrotron, etc.) or after absorbing neutrons in a nuclear reactor.

The elements thus transformed are called artificial radioactive isotopes. Of these, about 500 are known at present, of which close to 200 are already being used in practical applications. Apart from uranium 233 and the plutonium isotopes, which are discussed later, some of the most important are : hydrogen 3 (tritium), carbon 14, sodium 24, phosphorus 32, sulphur 35, potassium 42, calcium 45, chromium 51, iron 59, cobalt 60, krypton 85, strontium 90, yttrium 90, palladium 109, iodine 131 and 132, xenon 133, caesium 137, thulium 170, iridium 192, gold 198, and polonium 210.

Radioactive chemical elements and radioactive isotopes transform themselves naturally into more stable elements or isotopes.

The time required for the quantity of a given radioactive isotope to decrease to one-half that initially present is known as the half-life or transformation rate of that isotope. It varies from a fraction of a second for certain highly radioactive isotopes ($0.3 \times 10^{-6}$ for thorium C') to billions of years ($1.5 \times 10^{11}$ years for samarium 147) and constitutes a convenient yardstick of the statistical instability of the nucleus concerned.

Radioactive chemical elements and isotopes fall in this heading, even when mixed together or with radioactive compounds, or with non-radioactive materials (e.g., unprocessed irradiated targets and radioactive sources), provided that the specific radioactivity of the product is greater than 74 Bq/g (0.002 µCi/g).

(C) **Radioactive compounds; mixtures and residues containing radioactive substances**.

The radioactive chemical elements and isotopes of the present heading are often used in the form of compounds or products which are " labelled " (i.e., contain molecules with one or more radioactive atoms). Such compounds remain classified in this heading, even when dissolved or dispersed in, or mixed naturally or artificially with, other radioactive or non-radioactive materials. These elements and isotopes are also classified in this heading when in the form of alloys, dispersions or cermets.

Inorganic or organic compounds, chemically or otherwise constituted of radioactive chemical elements or radioactive isotopes, and solutions thereof, still fall in this heading, even if the specific radioactivity of these compounds or solutions is below 74 Bq/g (0.002 µCi/g); on the other hand, alloys, dispersions (including cermets), ceramic products and mixtures containing radioactive substances (elements, isotopes or compounds thereof) fall in this heading if their specific radioactivity is greater than 74 Bq/g (0.002 µCi/g). The radioactive elements and isotopes, very rarely used in their free form, are commercially available in chemical compounds or alloys. Apart from compounds of fissile and fertile chemical elements and isotopes, which are mentioned in Section (IV) below on account of their characteristics and importance, the most important radioactive compounds are :

(1) **Radium salts (chloride, bromide, sulphate, etc.**) used as radiation sources for treating cancer or for certain experiments in physics.

(2) **Compounds of radioactive isotopes referred to under (III) (B) above**.

Artificial radioactive isotopes and their compounds are used :

(a) **In industry**, e.g., for metal radiography, for measuring the thickness of sheets, plates, etc.; for measuring the level of liquids in an inaccessible container; for facilitating vulcanisation; to trigger off polymerisation or grafting of several organic compounds; for the manufacture of luminous paint (mixed, for example, with zinc sulphide); for clock and watch dials, instruments, etc.

(b) **In medicine**, e.g., for diagnosing or treating certain diseases (cobalt 60, iodine 131, gold 198, phosphorus 32, etc.).

(c) **In agriculture**, e.g., for sterilising agricultural produce; to prevent germination; for studies of fertiliser application or of fertiliser absorption by plants; to induce genetic mutations thus improving strains, etc. (cobalt 60, caesium 137, phosphorus 32, etc.).

(d) **In biology**, e.g., for studying the functioning or development of certain animal or vegetable organs (tritium, carbon 14, sodium 24, phosphorus 32, sulphur 35, potassium 42, calcium 45, iron 59, strontium 90, iodine 131, etc.).

(e) **In physical or chemical research**.

Radioactive isotopes and their compounds are normally put up in the form of powders, solutions, needles, thread or sheets. They are generally contained in glass ampoules, in hollow platinum needles, in stainless steel tubes, etc., which are packed in anti-radiation metal outer containers (generally of lead), the choice of thickness of which depends on the degree of radioactivity of the isotopes. In accordance with certain international agreements, a special label must then be affixed to the container, giving particulars of the isotope contained therein and its degree of radioactivity.

Mixtures may include certain neutron sources formed by associating (in a mixture, alloy, combinations, etc.) a radioactive element or isotope (radium, radon, antimony 124, americium 241, etc.) with another element (beryllium, fluorine, etc.) in such a way as to produce a (γ,n) or (α,n) reaction (introduction of a γ-photon or an α-particle, respectively, and emission of a neutron).

However, all assembled neutron sources, ready to be introduced into nuclear reactors to initiate a fission chain reaction, are to be considered as reactor components and consequently are to be classified in **heading 84.01**.

Microspheres of nuclear fuel coated with layers of carbon or silicon carbide intended for introduction into spherical or prismatic fuel elements fall in this heading.

Also included in this heading are the products used as luminophores, which have small quantities of radioactive substances added to make them self-luminescent, provided that the resulting specific radioactivity is greater than 74 Bq/g (0.002 µCi/g).

Of the radioactive residues, the most important from the point of view of re-use are :

(1) **Irradiated or tritiated heavy water** : after a residence time of varying length in a nuclear reactor, some of the deuterium in the heavy water is converted, by absorption of neutrons, into tritium and thus the heavy water becomes radioactive.

(2) **Spent** (irradiated) **fuel elements** (cartridges), generally very highly radioactive, mainly used for the purpose of recovering the fissile and fertile materials contained in them (see Section (IV) below).

## (IV) FISSILE AND FERTILE CHEMICAL ELEMENTS AND ISOTOPES AND COMPOUNDS THEREOF; MIXTURES AND RESIDUES CONTAINING THOSE SUBSTANCES

(A) **Fissile and fertile chemical elements and isotopes**.

Certain of the radioactive chemical elements and isotopes mentioned in Section (III) have a high atomic mass, for example thorium, uranium, plutonium and americium, of which the nucleus of the atom has a particularly complex structure. These nuclei, when subjected to the action of subatomic particles (neutrons, protons, deuterons, tritons, α particles, etc.) may absorb these particles, thereby increasing their instability to a degree sufficient to cause them to split into two nuclei of medium weight with neighbouring masses (or more rarely into three or four fragments). This disintegration liberates a considerable amount of energy and is accompanied by the formation of secondary neutrons. It is known as the process of **fission** or **nuclear bipartition**.

Fission only seldom occurs spontaneously or under the action of photons.

The secondary neutrons released at the time of fission may cause a second fission to take place thus creating secondary neutrons and so on. The repetition of this process produces a **chain reaction**.

The probability of fission is generally very high for certain nuclides (U 233, U 235, Pu 239) if slow neutrons are used, i.e., neutrons of an average speed of approximately 2,200 m/sec. (or an energy of 1/40 of an electron volt (eV)). As this speed corresponds approximately to that of the molecules of a fluid (thermal motion) slow neutrons are also sometimes called **thermal** neutrons.

At present, fission caused by thermal neutrons is that most used in nuclear reactors.

For this reason, the term **fissile** is commonly used to describe isotopes which undergo fission by thermal neutrons, particularly uranium 233, uranium 235, plutonium 239 and the chemical elements that contain them, particularly uranium and plutonium.

Other nuclides, such as uranium 238 and thorium 232 only undergo direct fission by fast neutrons and are commonly considered, not as fissile, but as **fertile**. The " fertility " comes from the fact that these nuclides can absorb slow neutrons, giving rise to the formation of plutonium 239 and uranium 233, respectively, which are fissile.

In thermal nuclear reactors (with slowed-down neutrons), since the energy of secondary neutrons released by fission is much higher (approximately 2 million eV), these neutrons have to be slowed down if a chain reaction is to take place. This can be achieved by means of **moderators**, i.e., products with a low atomic mass (such as water, heavy water, certain hydrocarbons, graphite, beryllium, etc.) which, although they absorb part of the energy of the neutrons by a succession of shocks, do not absorb the neutrons themselves or absorb only a negligible proportion of them.

In order to start and maintain a chain reaction, the average number of secondary neutrons produced by fission must more than compensate the neutrons lost by the phenomena of capture and escape not leading to fission.

The **fissile and fertile chemical elements** are listed below :

(1) **Natural uranium**.

Uranium in the natural state is composed of three isotopes : uranium 238, which forms 99.28 % of the total mass, uranium 235 which represents 0.71 %, and a negligible quantity (about 0.006 %) of uranium 234. Consequently, it can be considered as both a fissile element (because of its U 235 content) and a fertile element (because of its U 238 content).

Uranium is mainly extracted from pitchblende, uraninite, autunite, brannerite, carnotite or torbernite. It is also obtained from other secondary sources, such as residues from the manufacture of superphosphates or gold-mine waste. The normal process is reduction of the tetrafluoride by means of calcium or magnesium, or by electrolysis.

Uranium is a slightly radioactive element, very heavy (specific gravity 19) and hard. It has a lustrous silver-grey surface, but tarnishes on contact with the oxygen in the air, forming oxides. In powder form it oxidises and ignites rapidly when in contact with air.

Uranium is normally marketed in the form of ingots ready for polishing, filing, rolling, etc. (to produce bars and rods, tubes, sheets, wire, etc.).

(2) **Thorium**.

Since thorite and orangite, though very rich in thorium, are rare, thorium is mainly obtained from monazite which is also the source of rare-earth metals.

The impure metal takes the form of an extremely pyrophoric grey powder. It is obtained by electrolysis of the fluorides or by reduction of the fluorides, chlorides or oxides. The resulting metal is purified and sintered in an inert atmosphere and transformed into heavy steel-grey ingots (specific gravity 11.5); they are hard (although softer than uranium) and oxidise rapidly on contact with air.

These ingots are rolled, extruded or drawn to produce sheets, rods, tubes, wire, etc. Natural thorium consists essentially of the isotope thorium 232.

Thorium and certain thorium alloys are mainly used as fertile materials in nuclear reactors. Thorium-magnesium and thorium-tungsten alloys, however, are used in the aircraft industry or in the manufacture of thermionic devices.

Articles or parts of articles, made of thorium of Sections XVI to XIX are **excluded** from this heading.

(3) **Plutonium**.

Industrial plutonium is obtained by irradiating uranium 238 in a nuclear reactor.

It is very heavy (specific gravity 19.8), radioactive and highly toxic. It is similar to uranium in appearance, and in its oxidising propensities.

It is put up in the same commercial forms as enriched uranium and requires the greatest care in handling.

The fissile isotopes include :

(1) **Uranium 233** : this is obtained in nuclear reactors from thorium 232, which is transformed successively into thorium 233, protactinium 233 and uranium 233.

(2) **Uranium 235** : this is the only fissile uranium isotope which occurs in nature, being present in the proportion of 0.71 % in natural uranium.

To obtain uranium enriched in U 235 and uranium depleted in U 235 (enriched in U 238), uranium hexafluoride is submitted to isotopic separation by the electro-magnetic, centrifugal or gas-diffusion processes.

(3) **Plutonium 239** : this is obtained in nuclear reactors from uranium 238, which is successively transformed into uranium 239, neptunium 239 and plutonium 239.

Also to be mentioned are certain isotopes of transplutonium elements such as californium 252, americium 241, curium 242 and curium 244, which can give rise to fission (whether spontaneous or not) and which can be used as intense neutron sources.

Of the fertile isotopes, apart from thorium 232, depleted uranium (i.e. depleted in U 235 and consequently enriched in U 238) should be mentioned. This metal is a by-product of the production of uranium enriched in U 235. Because of its much lower cost and the large quantities available, it replaces natural uranium, especially as a fertile material, as a protective screen against radiations, as a heavy metal for the manufacture of fly-wheels or in the preparation of absorbent compositions (getters) used for purifying certain gases.

Articles or parts of articles, made of uranium depleted in U 235, of Sections XVI to XIX are **excluded** from this heading.

(B) **Compounds of fissile and fertile chemical elements or isotopes**.

The following compounds, in particular, fall in this heading :

(1) **of uranium** :

(a) the oxides $UO_2$, $U_3O_8$, and $UO_3$

(b) the fluorides $UF_4$ and $UF_6$ (the latter sublimes at 56 °C)

(c) the carbides UC and $UC_2$

(d) the uranates $Na_2U_2O_7$ and $(NH_4)U_2O_7$

(e) uranyl nitrate $UO_2(NO_3)_2.6\ H_2O$

(f) uranyl sulphate $UO_2SO_4.\ 3\ H_2O$

(2) **of plutonium** :

(a) the tetrafluoride $PuF_4$

(b) the dioxide $PuO_2$

(c) the nitrate $PuO_2(NO_3)_2$

(d) the carbides PuC and $Pu_2C_3$

(e) the nitride PuN.

The uranium or plutonium compounds are mainly used in the nuclear industry, either as intermediates or as finished products. The uranium hexafluoride is usually presented in sealed containers; it is rather toxic and should therefore be handled with care.

(3) **of thorium** :

(a) oxide and hydroxide. Thorium oxide ($ThO_2$) (thoria) is a whitish-yellow powder, insoluble in water. The hydroxide ($Th(OH)_4$) is hydrated thoria. Both are obtained from monazite. They are used in the manufacture of gas-mantles, as refractory products or as catalysts (acetone synthesis). The oxide is used as fertile material in nuclear reactors;

(b) inorganic salts. These salts are usually white. The most important are :

(i) thorium nitrate, appearing in the more or less hydrated state as crystals, or as powder (calcined nitrate). It is used to prepare luminescent paints. Mixed with cerium nitrate it is used to impregnate gas-mantles;

(ii) thorium sulphate, a crystalline powder, soluble in cold water; thorium hydrogen sulphate and alkali double sulphates;

(iii) thorium chloride ($ThC1_4$), anhydrous or hydrated, and oxychloride;

(iv) thorium nitride and thorium carbide. Used as refractory products, as abrasives or as fertile materials in nuclear reactors;

(c) organic compounds. The best known organic compounds are thorium formate, acetate, tartrate and benzoate, all used in medicine.

(C) **Alloys, dispersions (including cermets), ceramic products, mixtures and residues containing fissile or fertile elements or isotopes or inorganic or organic compounds thereof**.

The principal products in this group are :

(1) **Alloys of uranium or plutonium with** aluminium, chromium, zirconium, molybdenum, titanium, niobium or vanadium. Also uranium-plutonium and ferro-uranium alloys.

(2) **Dispersions of uranium dioxide** ($UO_2$) **or of uranium carbide** (UC) (whether or not mixed with thorium dioxide or thorium carbide) in graphite or polyethylene.

(3) **Cermets** consisting of various metals (e.g. stainless steel) together with uranium dioxide ($UO_2$) plutonium dioxide ($PuO_2$) uranium carbide (UC) or plutonium carbide (PuC) (or these compounds mixed with thorium oxide or carbide).

These products in the form of bars, plates, spheres, threads, powder, etc., are used either for the manufacture of fuel elements or, sometimes, directly in the reactors.

Bars, plates and spheres, contained in a sheath and fitted with special attachments for handling purposes, fall in **heading 84.01**.

(4) Spent or irradiated fuel elements (cartridges), that is, those which, after more or less extensive use, must be replaced (e.g., because the accumulation of fission products is hampering the chain reaction or because the sheath has deteriorated). After sufficiently long storage in very deep water to cool them and to allow their radioactivity to decrease, these fuel elements are transported in lead containers, to specialised installations equipped for the recovery of the residual fissile material, of the fissile material derived from the transformation or fertile elements (which are usually contained in fuel elements) and of fission products.