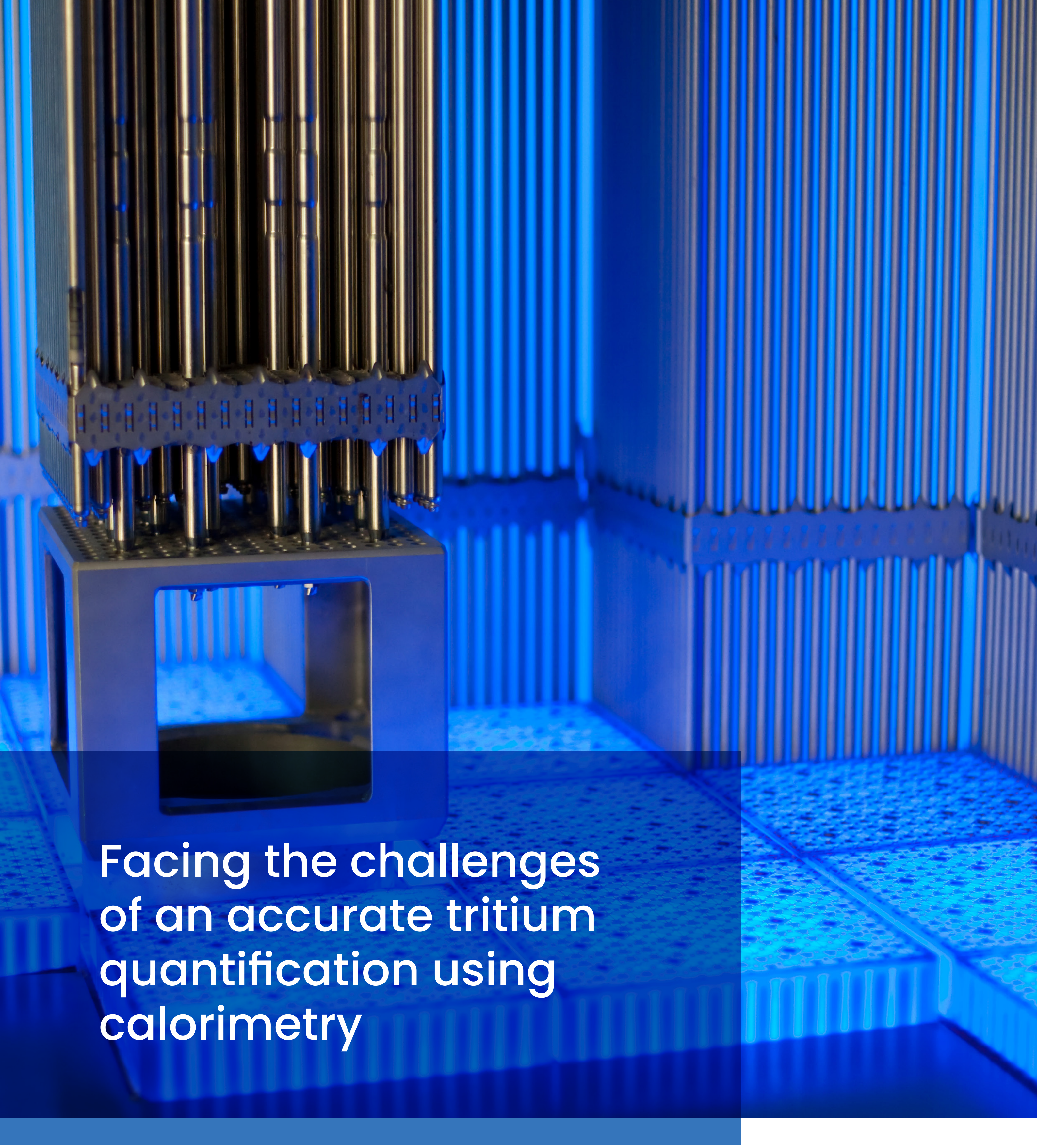

Facing the challenges of an accurate tritium quantification using calorimetry

setsafe
KEP TECHNOLOGIES

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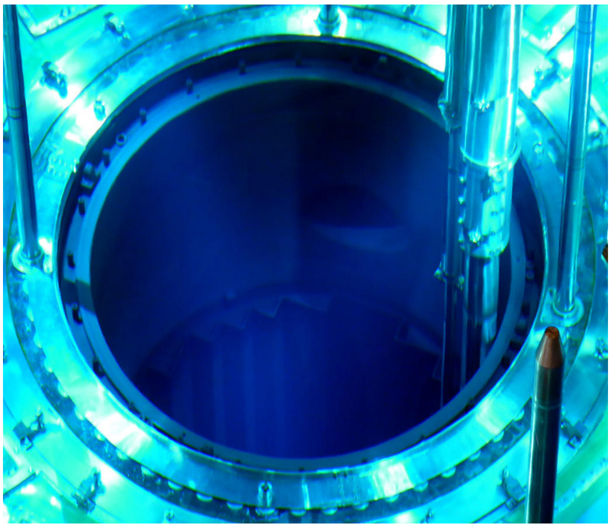
1.

Why characterize and quantify nuclear materials and waste?



For any nuclear facility, managing inventories or quantities of radioactive materials and waste is a major challenge, with multiple aspects: protection of workers and the population against ionizing radiation hazards, valorization of this inventory through economic development, risk mitigation of loss, theft, or detour of these materials and waste for hostile purposes, and limitation of the constraints imposed on future generations.

To tackle some of these challenges, national and international regulations require inventories, i.e. material balances, to be carried out at different scales.

For instance, circular 153 of the International Agency for Atomic Energy indicates that agreements between Member States and AIEA must « *provide that the State shall establish and maintain a system of accounting for and control of all nuclear material subject to safeguards [...] and that such safeguards shall be applied [...] in ascertaining that there has been no diversion of nuclear material from peaceful uses to nuclear weapons or other nuclear explosive devices.” Moreover, “the agreement should provide that the State's system of accounting for and control [...] shall be based on a structure of material balance areas, and shall make provision [...] of such measures as: (a) A measurement system for the determination of the quantities of nuclear material received, produced, shipped, lost or otherwise removed from inventory, and the quantities on inventory; (b) The evaluation of precision and accuracy of measurements and the estimation of measurement uncertainty”* .[1]

These facilities must therefore apply simple and safe solutions to rigorously control the quantity of materials on their sites using reliable characterization means.

Tritium is produced in large quantities in pressurized heavy water reactors (PHWR), the majority of which are of the CANDU (CANadian Deuterium natural Uranium) type.

2.

Tritium, its sources and uses

Tritium is a radioactive isotope of hydrogen that disintegrates into the stable helium 3, with a 12.33 years half-life.

The main tritium sources

Tritium is produced naturally and is also a by-product of the nuclear industry. It exists in its gaseous form, but can also be found in oxidized form (tritiated water or water vapor) or organic form (bound to carbon).

The action of cosmic rays (neutrons) on certain elements in the air (nitrogen 14) leads to a natural production of atmospheric tritium of between 0.15 to 0.20 kg per year. It is also produced in the earth's crust, but in very small quantities [2].

Most nuclear reactor technologies produce tritium. Some generate such small quantities that it is neither worth recovering nor dangerous to be released into the environment.

This is true for direct releases in gaseous or liquid form, or through fuel processing plants. On the other hand, tritium is produced in large quantities in pressurized heavy water reactors (PHWRs), the majority of which are of the CANDU type (for CANadian Deuterium natural Uranium) [3]. It is mainly in the form of tritiated water (HTO) and can be extracted for industrial use.

This process is applied in Tritium Removal Facility (TRF) in two stages: catalytic extraction in vapor phase, then cryogenic distillation for an approximate production of a few hundred grams per year and per facility [3] [4] [5].

Main uses of tritium

The thermonuclear fusion reaction between deuterium and tritium is the basis of the nuclear deterrence arsenal of some countries [6]. But this reaction will also be exploited in nuclear fusion reactors.

This is in particular the purpose of the research undertaken on an international scale with the ITER project.

Tritium needs are significant in this sector, with an estimated 25 kg for the ITER experimental device. For the next step, the DEMO reactor, which should bring fusion to the threshold of industrial operation, tritium requirements will be of the order of 300 g per day to produce 800 MW of electricity [7].

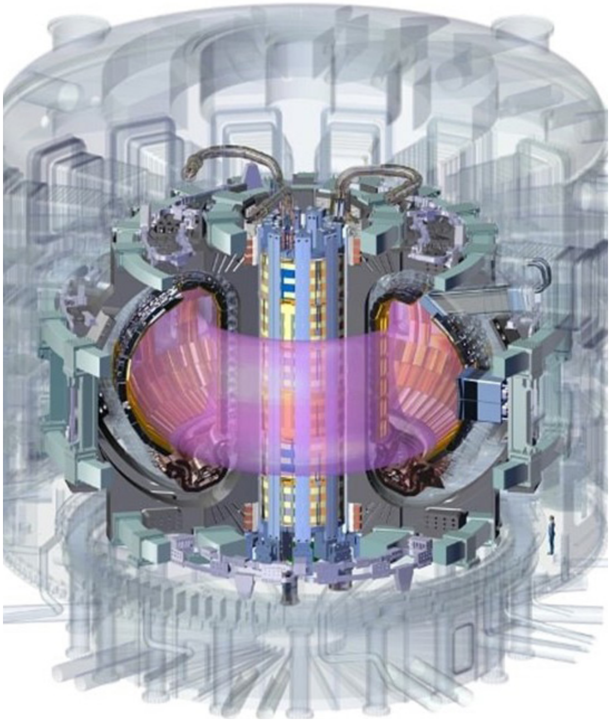


Figure – The central solenoid of the ITER tokamak, an experimental device designed to exploit the energy of deuterium and tritium fusion
(Credit © ITER Organization, http://www.iter.org/)

More peripheral applications use tritium's properties to make phosphorescent materials glow for light signaling, or use it as a radiochemical product or as a tracer for research or oil and gas exploitation.

3.

Tritium quantification: challenges and opportunities

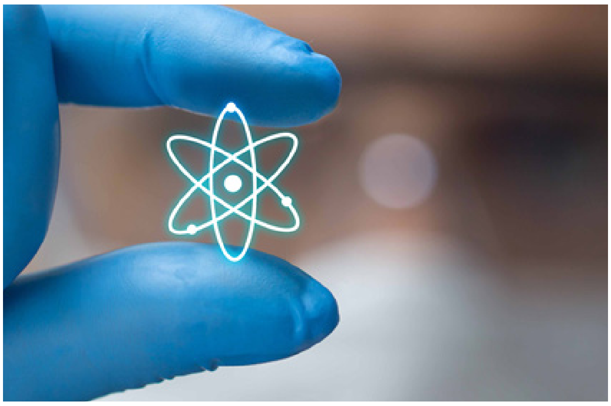
Tritium accounting and inventories, tritium waste management

The monitoring needs in the context of radioactive material and waste disposal are detailed in this document's introduction. But the challenges faced by tritiated material and waste characterization are numerous.

On the one hand, its storage and disposal are made difficult because of its tendency to outgas. Tritium from military installations is notably the source of tritiated waste that poses problems, more because of tritium's mobility than because of its radioactive toxicity [6].

On the other hand, the concentration of tritium is very variable within the same component. Destructive measurements by sampling are therefore not very representative.

Apart from liquid scintillation dedicated to low concentrations, conventional nuclear measurement techniques are not fully adapted to tritium measurement and only chemical analysis techniques meet this need [8]. However, they remain sampling-based techniques, with the above mentioned problems of representativeness.



Tritium quantification for commercial exchange

Since tritium is produced worldwide in very small quantities (as a reminder, a CANDU reactor produces a few hundred grams of tritium per year), and since its extraction process is complex and therefore expensive, a gram of tritium is traded at a high price, from about $30,000 to about $35,000 [9] [10]. Therefore, a facility produces the equivalent of a few million to a few tens of millions of US dollars a year.

The measurement accuracy of the tritium quantities exchanged between the producer and the user is therefore critical because it has a direct economic impact. The slightest measurement uncertainty would result in an under- or over-estimation that would strongly disadvantage one party or the other.

Tritium quantification for its transportation

Compared to storage, the transport of radioactive materials presents increased risks, particularly in terms of loss, theft or misappropriation. Thus, international regulations govern the transport of these materials and require the measurement of their quantity (or activity).

IAEA indicates for example that « *the material to be transported should be characterized to identify the radionuclides, the form and activities of the material in order to assign a transport security level. In some cases, a shipment might consist of a single radionuclide, either in a single package or multiple packages. In other cases, there might be multiple radionuclides within a single package or multiple packages containing multiple radionuclides within a single shipment. The identity and activity level of each of the radionuclides should be identified* » [11]

A shipment containing tritium must therefore be quantified in order to assign the appropriate level of transport security.

Tritium storage, today highly centralized on specific sites, combined with increasing commercial exchanges, will thus impose the need for accurate measurements on all shipping sites and all recipient sites.

4.

Calorimetry as a technique for measurement and quantification of tritium

Calorimetry is a technique based on the measurement of heat released by a package of radioactive material or waste. It is known to be very reliable, particularly for characterizing tritium or plutonium.

It has indeed interesting advantages for these measurements:

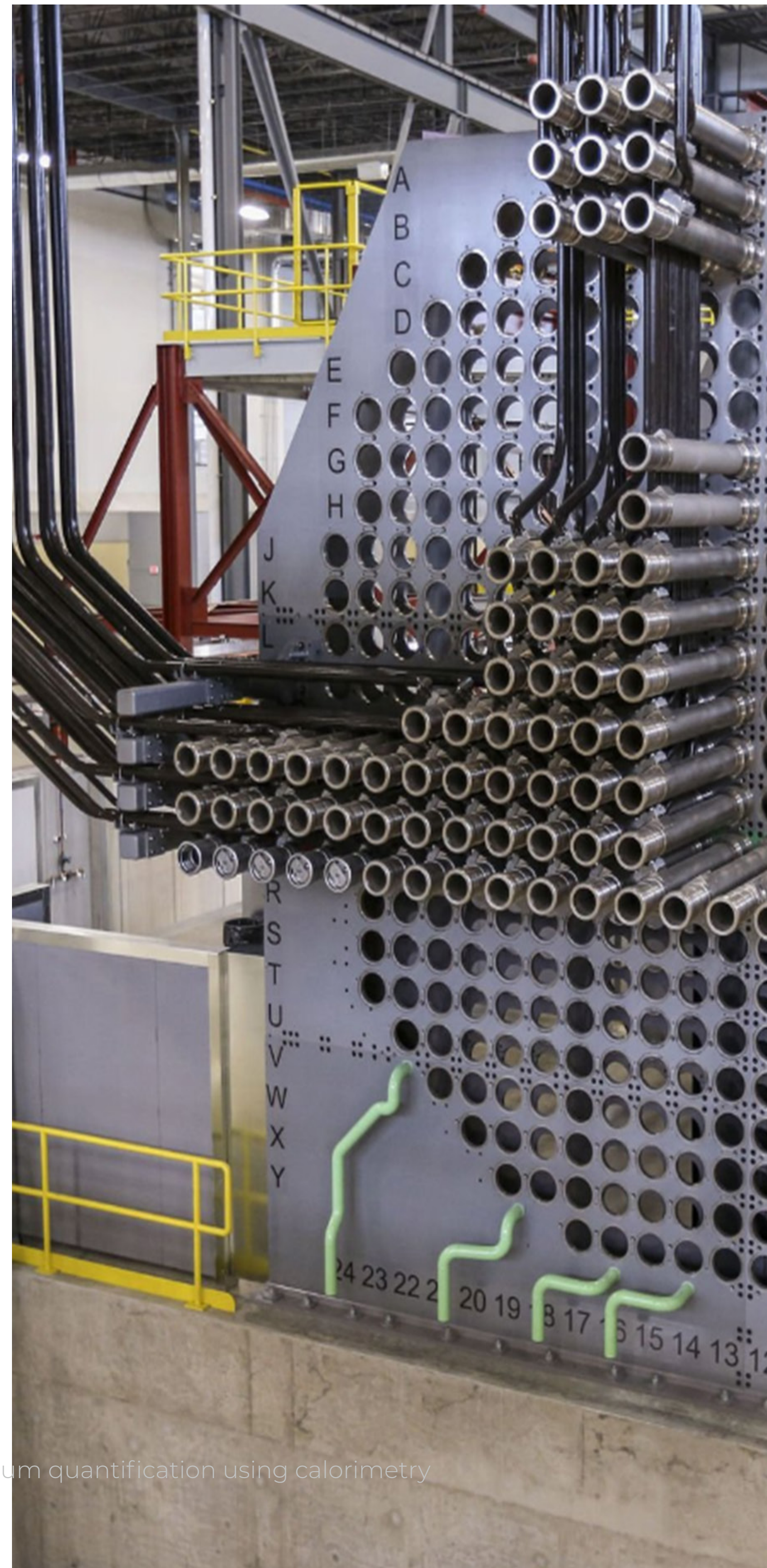
1. The measurement is non-destructive and performed on the whole container, so it is representative.

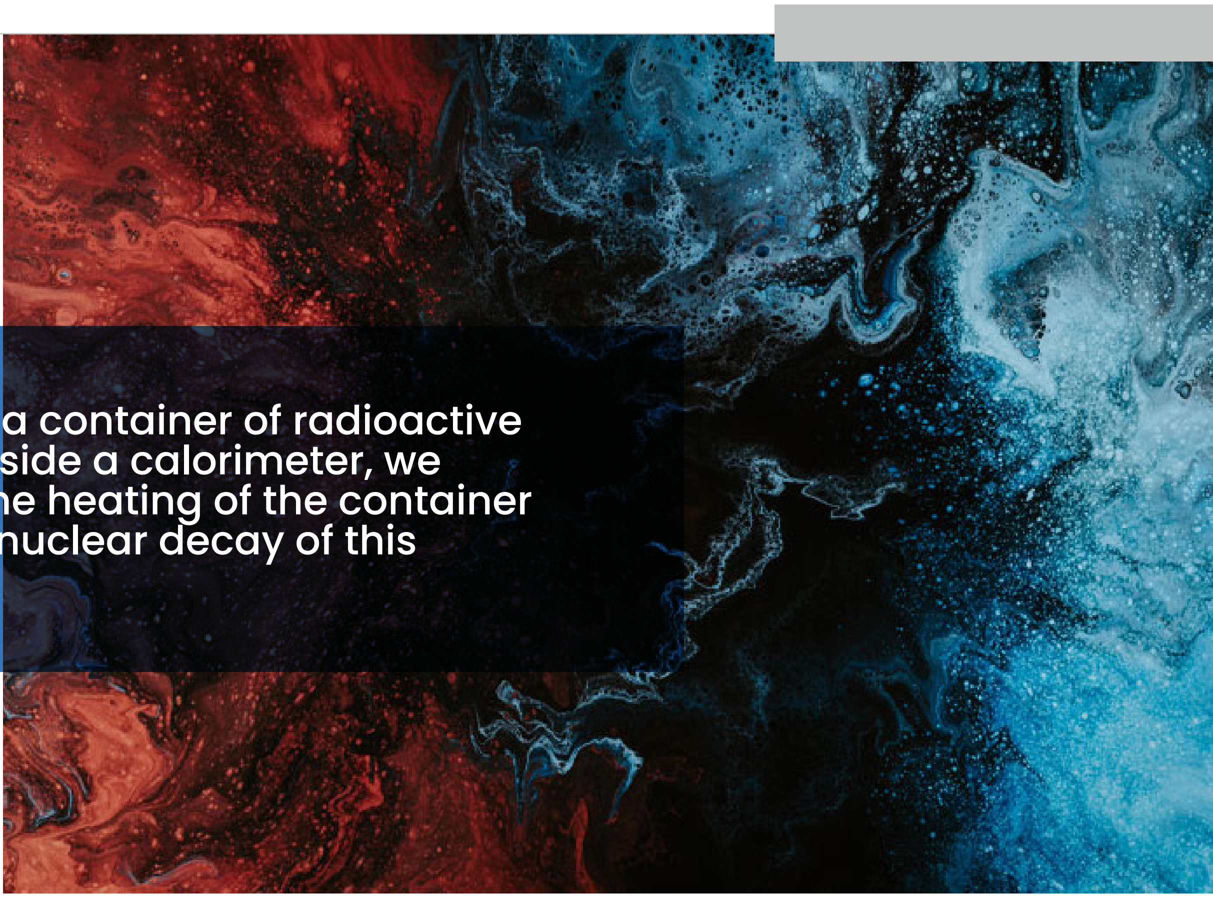
2. Calorimetry allows measurement accuracies of tritium quantities better than 0.5% [12], and is therefore considered to be the most reliable of the non-destructive tritium analysis techniques.

3. Calorimetry can be used on any type of matrix or material because its measurement results are not influenced by them [13].

4. The calorimetric measurement does not depend on the geometry of the object or on the chemical form of the radioelement [8].

The principle of calorimetry measurements and its use for tritium quantification are detailed in the following chapters.





By placing a container of radioactive material inside a calorimeter, we measure the heating of the container due to the nuclear decay of this material.

5.

The general principle of calorimetry to characterize nuclear material and waste

The nuclear decay of radioactive elements generates charged particles. These particles are slowed down and then stopped by passing through surrounding materials. In doing so, they deposit some of their energy, causing the materials to heat up.

Calorimetry is a method to detect and measure the heat exchanged between an object and its environment. Thus, by placing a container of radioactive material inside a calorimeter, the container's heat flow due to nuclear decay of the material is measured.

If the container is designed with a material dense and thick enough to stop all the charged particles, and in the absence of any other heat source, there is a linear relationship between the thermal power measured by the calorimeter and the activity of the radioactive material.

The heat flow released by the container is thus directly proportional to the amount of material it contains, and a calorimetric measurement becomes a measurement of the amount of radioactive material.

Calorimeters for the characterization of radioactive material or waste containers are called "isothermal", because they regulate the temperature of the container environment so that it is stable enough not to disturb the heat flow measurement. The control setpoint is set by the user within the operating range of the calorimeter.

Most calorimeters use the principle of differential heat flow measurement based on a specific design using two perfectly identical twin cells. One is called the measuring cell and contains the package to be characterized, the other is called the reference cell and generally contains an empty, thermally inert container. This concept provides a better stabilization of the calorimetric signals and improves the measurement performance.

There are exceptions, in which the reference cell is replaced by a much smaller version (Ghost Cell), but with the same function of stabilizing the calorimetric signal. This approach is used in very large volume calorimeters in order to limit the equipment’s footprint.

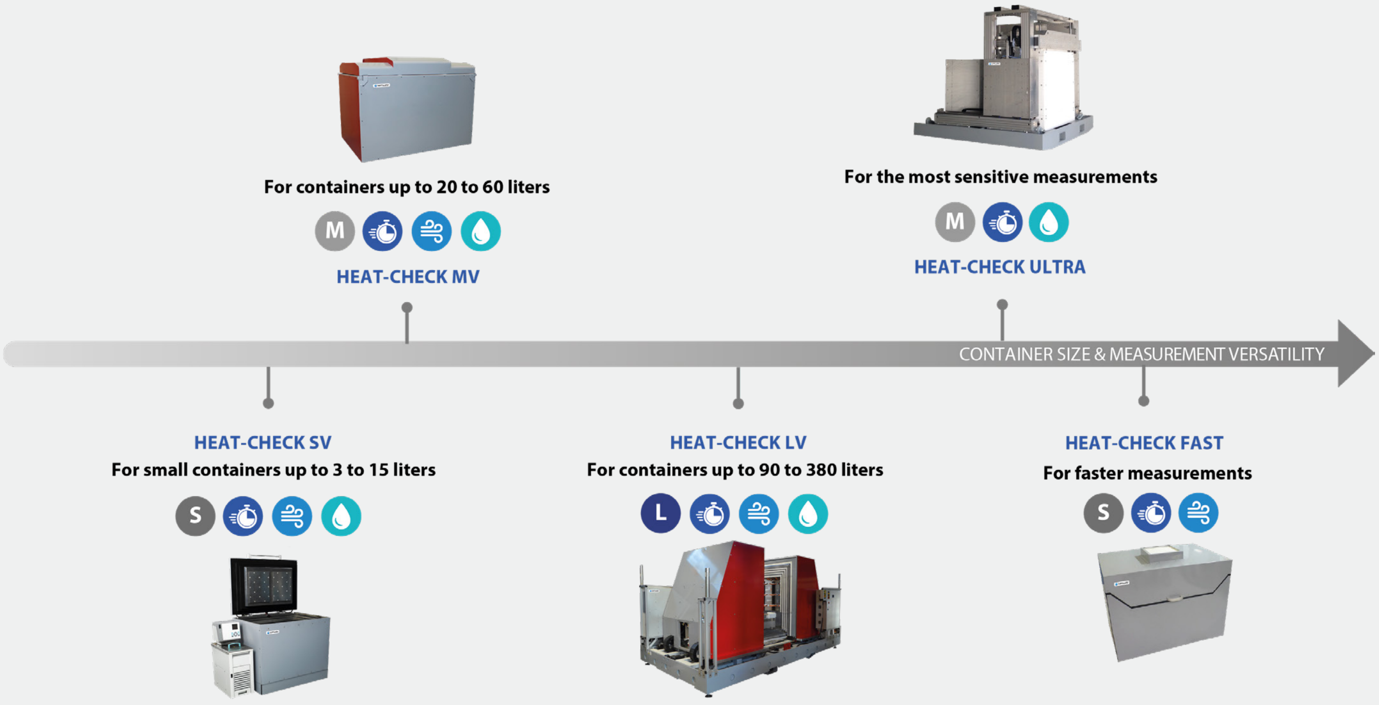


Figure – Representation of Setsafe’s range of calorimeters. The models differ in terms of capacity, measurable containers' volume, detection limits and measurement time.

The size of the cells in each calorimeter is optimized for the volumes of the containers to be characterized, from a few liters to a few hundred liters.

The heat flow sensors that equip these cells use thermoelectric elements, which generate an electrical signal when they detect heating. Thus, to display a heat flow signal instead of an electrical one, calibration is necessary.

This is achieved with the help of so-called "Joule effect" devices. Basically, these are very precise heating resistors, which release a heat flow set by the operator.

The response of the calorimeter signal to this Joule effect is analyzed and is used to calibrate its heat flow measurement, before carrying out series of measurements on containers.

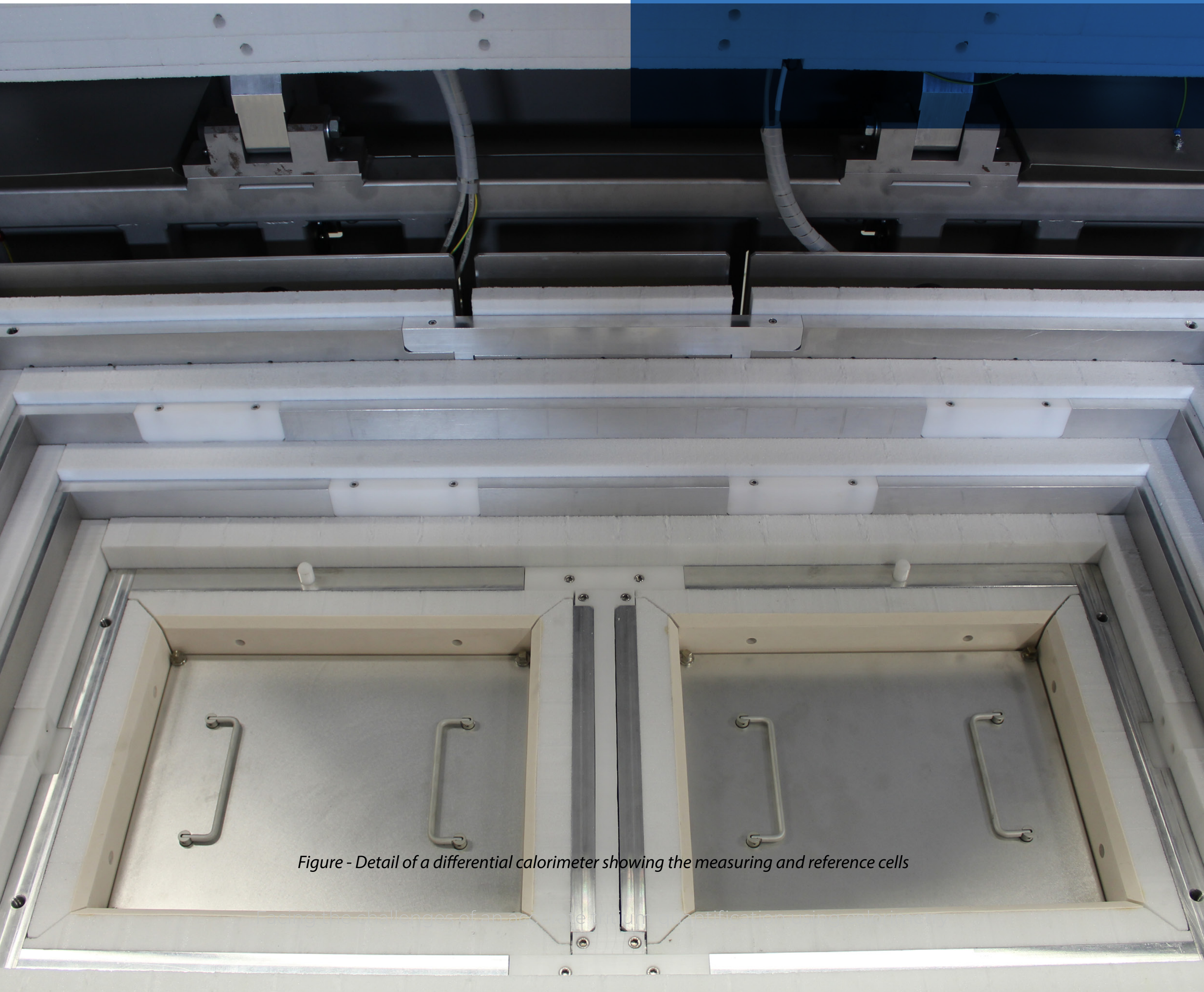


Figure - Detail of a differential calorimeter showing the measuring and reference cells

6.

The specific principle of tritium measurement by calorimetry

Tritium emits low-energy beta radiation (Emax = 18.6 keV, Emoy = 5.7 keV) that can be stopped by a very small thickness of material.

Most containers of tritiated products are therefore capable of collecting the entire energy deposited by tritium beta radiation. In the absence of any other heat source, the thermal power released by a tritium container is thus directly proportional to the amount of material it contains.

The physical quantity that relates this heat release to a radioactive material quantity is called the specific power. The specific power of tritium is equal to 324 milliwatts per gram of tritium.

The calorimeter, after calibration, measures a heat flow generated by the package measured in milliwatts. The amount of tritium in the package is then simply calculated from the following equation:

$$Mass_{tritium} = \frac{Heat\ flow_{container}}{Specific\ power_{tritium}}$$

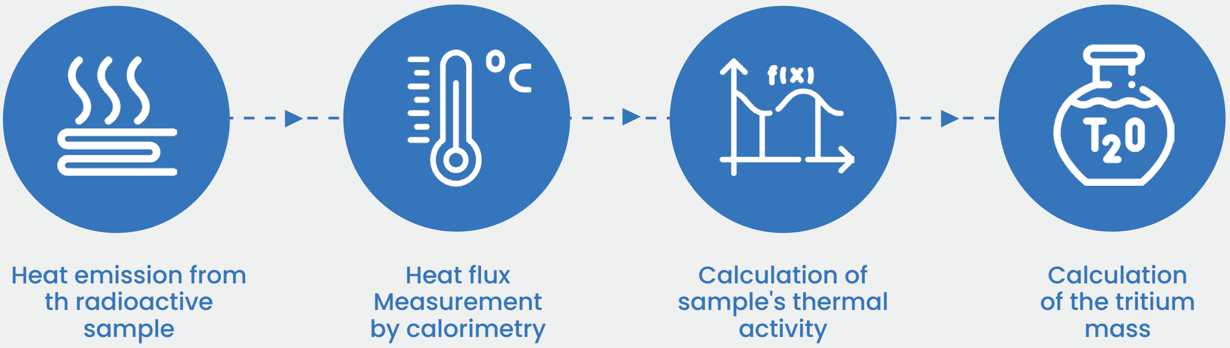


Figure - Principle of measurement by calorimetry

A tritium container with a measured heat output of 32.4 milliwatts therefore contains 0.1 grams of tritium. Typically, calorimetry is applied to measurements of tritium quantities from 1 milligram to 400 grams [14].

The activity is calculated as follows:

$$Activity_{tritium} = \frac{Heat\ flow_{container}}{Specific\ power_{tritium}} \times Activity\ mass_{tritium}$$

The mass activity being equal to: 356TBq/g.

There are four simple steps to the measurement:

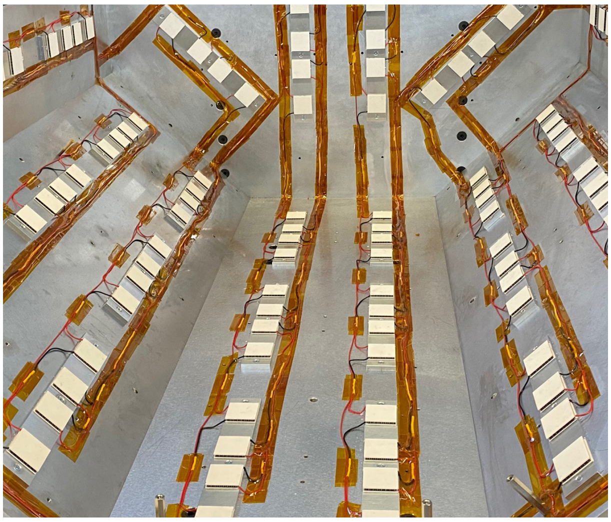
1. The container is introduced into the calorimeter and the latter is closed to avoid heat losses to the outside,

2. The temperature and heat exchange are stabilized for a period of time depending on the characteristics of the container and of the calorimeter,

3. The heat flow generated is plotted from the calorimeter signal,

4. This heat flow value is converted into tritium quantity using the relation seen above.

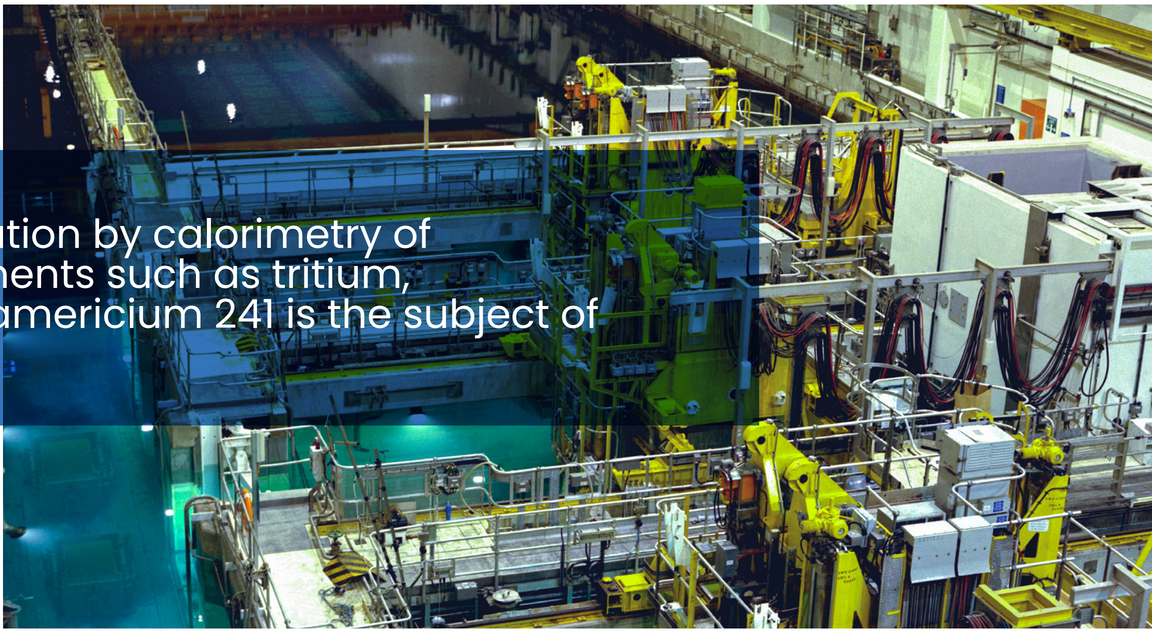
Among many practical applications of this method around the world, we can cite the CEA in France, which quantifies, among other things, the tritiated water adsorbed on a zeolite inside molecular sieve traps [15].



In Korea, KEPRI uses calorimetry to control the quantities of tritium coming from its WTRF (Wolsong Tritium Removal Facility) and supplied to its customers (ITER, industries, etc) [12]. EFDA (now EUROfusion) uses calorimetry to monitor and characterize the efficiency of the Exhaust Detritiation System (EDS) of the JET (Joint European Torus) research facility [16].

More generally, calorimetry is considered to be an excellent addition to conventional non-destructive testing techniques in nuclear measurement. Its use in the measurement of tritium is not limited: it is also widely used for the quantification of radioelements such as americium, strontium or plutonium [8].

The ASTM C1458-16 standard [17] mentions, for example, the measurement of plutonium in mixed oxides of uranium and plutonium (PuO_2-UO_2), the measurement of plutonium in molten salt residues, and the measurement of americium 241 in molten salt residues.



The characterization by calorimetry of radioactive elements such as tritium, plutonium, and americium 241 is the subject of a standard.

7.

Our references

Users and testimonials

« *Calorimetry is the most reliable of the non-destructive analytical techniques for the quantification of plutonium or tritium contents in a matrix* » [13].

« *Calorimetry is a non-destructive method that can be used successfully to measure tritium and provides high accuracy in static measurements* ». [5]

« *In general, calorimetry has many advantages: it is non-intrusive, non-destructive, does not depend on the geometry of the object or the chemical form of the radioactive element, and requires no sampling or sample preparation, which makes it particularly attractive for quantifying nuclear material inventories. The radioactive half-life of tritium (12.3 years) is large compared to the measurement times (a few days), making calorimetry a perfectly suitable technique for quantifying tritium* » [8]

Standards

Calorimetric characterization of radioactive elements such as tritium, plutonium, and americium-241 has been the subject of an American Society for Testing and Materials standard since 2000.

This standard has been revised several times and its latest version was published in December 2016:
ASTM C1458-16, Standard Test Method for Non-destructive Assay of Plutonium, Tritium and 241Am by Calorimetric Assay [Report]. - West Conshohocken, United States : ASTM International, 2016.

8.

About us

KEP Technologies

KEP Technologies is a family-owned industrial group created in 1996. KEP Technologies is a medium-sized companymore than 600 employees and a turnover of more than 70 M€.

With an international orientation through its industrial and commercial presence on 5 continents, it offers its customers industrial and technological solutions in a wide range of markets: aeronautics, space, health, research, defense, security, energy, environment, transport, luxury...

The KEP Technologies group, through its various brands, skills and activities, supports its customers in the nuclear sector to meet their challenges.

These require the use of specific solutions that involve expertise, innovation capabilities, diversified skills, certifications (CEFRI) and mastery of project management methods.

Setsafe

Through its Setsafe brand, KEP Technologies offers solutions for the measurement of radioactivity and its implementation in the nuclear sector.

We work with our customers to provide standard or customized solutions to meet the challenges related to the measurement of radioactivity.

Our expertise

Our team of experts can lead the project from design to installation, training and maintenance, or any other need. Our expertise in engineering, industrial control and metal systems allows us to intervene on vast and varied projects, while respecting all the constraints of the nuclear sector.

KEP Technologies holds two patents concerning calorimetry for the characterization of radioactive materials and waste:

☑ EP2946184 DIFFERENTIAL CALORIMETER FOR HEAT FLOW MEASUREMENT in co-ownership with the French Atomic Energy and Alternative Energies Commission.

☑ FR3028948 CALORIMETER FOR MEASURING A QUANTITY OF ACTIVE MATERIAL with the French Atomic Energy and Alternative Energies Commission.

9.

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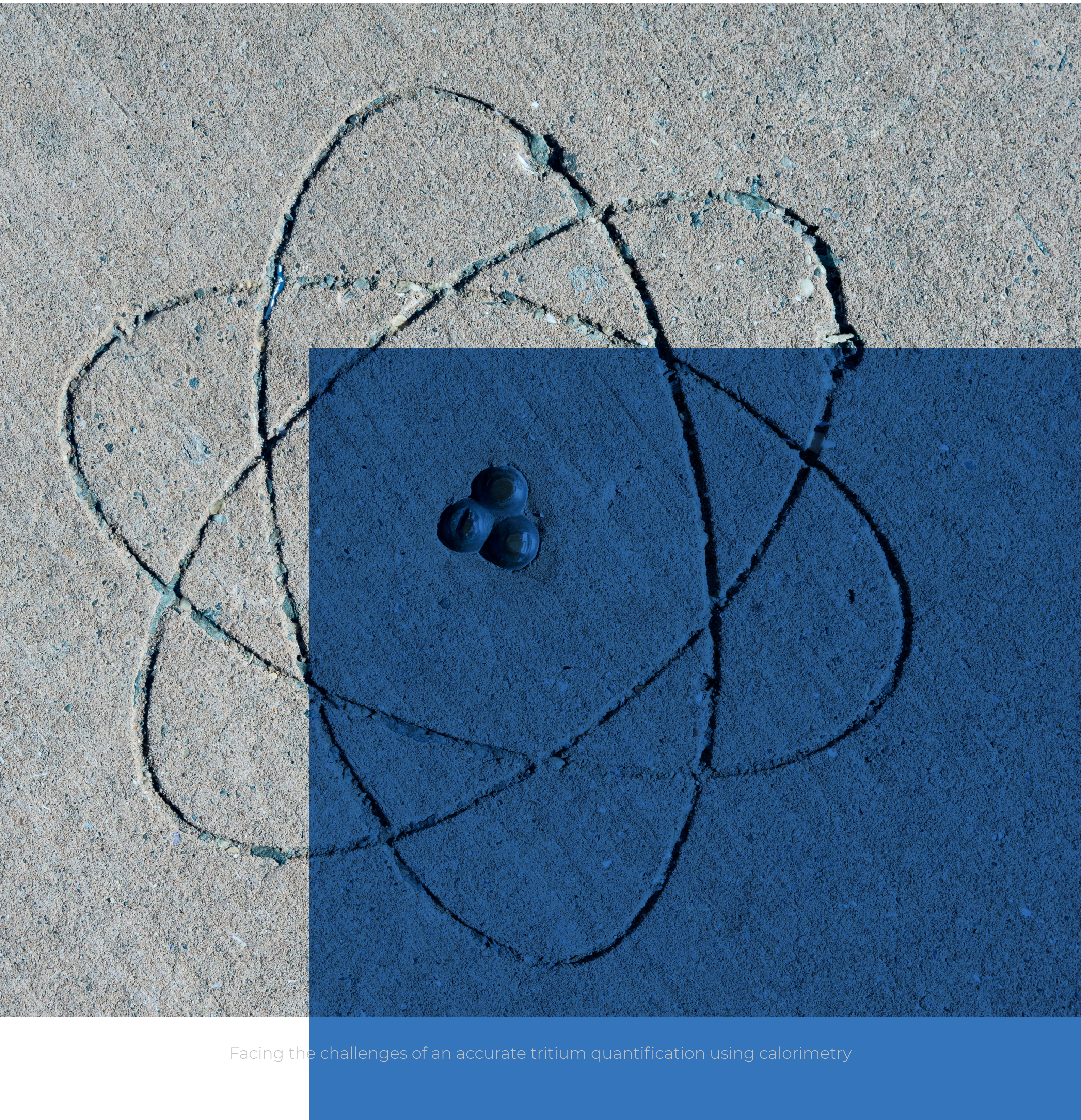

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