INTERNATIONAL STANDARD

ISO 16659-1

First edition
2022-12

Ventilation systems for nuclear facilities — In-situ efficiency test methods for iodine traps with solid sorbent —

Part 1: General requirements

Systèmes de ventilation pour les installations nucléaires — Méthodes d'essai in-situ de l'efficacité des pièges à iode à sorbant solide —

Partie 1: Exigences générales

Reference number
ISO 16659-1:2022(E)

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Published in Switzerland

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Foreword

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This document was prepared by Technical Committee ISO/TC 85, *Nuclear energy, nuclear technologies, and radiological protection*, Subcommittee SC 2, *Radiological protection*.

A list of all parts in the ISO 16659 series can be found on the ISO website.

Any feedback or questions on this document should be directed to the user's national standards body. A complete listing of these bodies can be found at www.iso.org/members.html.

Introduction

In nuclear facilities, iodine traps are usually used on ventilation systems to limit radioactive iodine effluent releases into the environment, to reduce iodine concentration in the air of facilities by recycling or to prevent radioactive iodine from entering into protected areas (such as control room for example). Some examples of the iodine trapping systems are shown in Annex B. The knowledge or the warranty of the capacity of these devices to trap iodine could be necessary, particularly when they are valued in the safety demonstration.

The IAEA recommends in the Safety Guide SSG-53[21] to test periodically the efficiency of confinement systems used to limit gaseous radioactive effluents releases into the environment. This recommendation is transcribed in some national rules by requirements about testing the efficiency of filtration or scrubbing devices of facilities' ventilation systems but, no international standard exists for the methods to be used for testing them in situ. ISO 17873 and ISO 26802 recommend periodic testing after their installation as well. Some design recommendations may also be found in national standards (e.g. ASTM standard[8]).

This document is the general part of a set of standards on the different current methods of tests. It describes common provisions to use to test in situ the iodine trap scrubbing efficiency of ventilation systems of nuclear facilities. These provisions deal with the methods used according to the expected role of this iodine trap, requirements about workers protections, and requirements for environment protection to take into account during these tests. Specific methods will be presented in the different parts of ISO 16659, using radioactive nuclides (e.g. $^{131}ICH_3$ in order to determine the filters efficiency or gases such as cyclohexane in order to perform integrity tests).

Ventilation systems for nuclear facilities — In-situ efficiency test methods for iodine traps with solid sorbent —

Part 1: General requirements

1 Scope

The scope of ISO 16659 series is to provide different test methods aiming at assessing the efficiency of radioactive iodine traps in ventilation systems of nuclear facilities. The ISO 16659 series deals with iodine traps containing a solid sorbent — mainly activated and impregnated charcoal, the most common solid iodine sorbents used in the ventilation systems of nuclear facilities — as well as other sorbents for special conditions (e.g. high temperature zeolites).

The scope of this document is to provide general and common requirements for the different test methods for industrial nuclear facilities. The different methods will be described in other specific parts of ISO 16659 series. Nuclear medicine applications are excluded from the scope of ISO 16659 series.

In principle, ISO 16659 series is used mainly for filtering radioactive iodine, but other radioactive gases can also be trapped together with iodine. In such a case, some specificity may have to be adapted for these other radioactive gases in specific parts of ISO 16659 series.

This document describes the main general requirements in order to check in situ the efficiency of the iodine traps, according to test conditions that are proposed to be as reproducible as possible.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 2889:2021, *Sampling airborne radioactive materials from the stacks and ducts of nuclear facilities*

3 Terms and definitions

For the purposes of this document, the following terms and definitions apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at https://www.electropedia.org/

3.1
activated and impregnated charcoal
carbon fiber filter
charcoal or carbon fiber filters often obtained from biomass or synthetic fiber precursors and for which its specific surface is drastically increased by physical or chemical activation during a high temperature thermal treatment

Note 1 to entry: Its specific surface area is so high and so its adsorption capacity, that it is largely used in iodine trap in nuclear installations or mask for workers.

Note 2 to entry: The activated charcoal can be impregnated with potassium iodide (KI) and/or triethylenediamine (TEDA) to enhance the decontamination factor by increasing respectively isotopic exchange and chemical adsorption.

Note 3 to entry: Activated carbon fiber filter are also used for nuclear applications for iodine trap in nuclear installations or mask for workers; their principle is to use microporous trapping phenomena (increasing the specific surface area) instead of macro-porous trapping phenomena in conventional charcoal filters.

3.2
adsorption
surface phenomenon that results in the increase of the density of an adsorbate, a substance that is adsorbed (atoms, ions or molecules from a gas, liquid or dissolved solid), due to fixation to a surface by different physical (physisorption) or chemical (chemisorption) processes with different energies

Note 1 to entry: It differs from absorption, in which a fluid (the absorbate) is dissolved by or permeates a liquid or solid (the absorbent), respectively. Absorption is a volume phenomenon.

Note 2 to entry: Charcoal, argil or zeolite are good adsorbent due to their crystalline structure.

3.3
chemical adsorption
chemisorption
adsorption (3.2) resulting from a surface chemical reaction between the adsorbate and the impregnated surface of the sorbent with formation of a chemical bond

Note 1 to entry: This irreversible phenomenon leads to a deep modification of the repartition of the electronic charges of the adsorbed molecules, the forces are similar to the chemical bond.

3.4
contact time
gas flow transit time through sorbent layer (or sorbent bed)

Note 1 to entry: Contact time, τ, is expressed using the ratio: sorbent thickness (in metres)/frontal speed (in meter per second) or using the ratio: sorbent volume (in cubic meter)/flow rate throw the sorbent (in cubic metres per second).

3.5
decontamination factor
f_D
measure of the efficiency achieved by a filter and corresponding to the ratio of activity, A, of the species, expressed in Bq at the inlet of the filter (or concentration of tracer, $C_{upstream}$) and the activity of the species in Bq, a, (or concentration of tracer, $C_{downstream}$) at the outlet of the filter

Decontamination factor f_D is expressed using the following formula:

$$f_D = A/a = C_{upstream}/C_{downstream}$$

where A and a are the activity upstream and downstream and $C_{upstream}$ and $C_{downstream}$ are the concentration upstream and downstream, the f_D being greater than 1.

Note 1 to entry: The decontamination factor is related to efficiency, E, and penetration, P, by the following relation:

$$f_D = \frac{1}{1-E} = \frac{1}{P}$$

Note 2 to entry: The decontamination factor considers both the intrinsic quality of the sorbent and leaks of the in situ complete integrated device (internal, due to mounting, by-pass, etc.).

Note 3 to entry: The notion of decontamination factor applies particularly to tests with radioactive tracer gas.

3.6
desorption
inverse phenomenon of the physical *adsorption* (3.2) due to physical or chemical modification of the sorbent (increase of temperature, decrease of pressure, etc.)

3.7
efficiency
E
ratio of the quantity of species (particles or gas) retained by the filter to the quantity entering it

Note 1 to entry: Efficiency is always less than or equal to 1.

3.8
frontal speed
speed of gaseous radioactive wastes through sorbent bed layer

3.9
hygrometric and thermal equilibrium
conditions for which hygrometric or thermal parameters of the air flow containing water vapor crossing the sorbent start to reach asymptotic value downstream the sorbent such as it can be considered that the sorbent and air flow are in equilibrium

3.10
integrity test
in situ test indicating whether the filter or material is performing as designed, such as to identify potential non-filtered leaks

3.11
iodine sorbent
sorbent intended for trapping radioiodine in gaseous radioactive effluent, usually based on activated and impregnated charcoal, silver impregnated zeolite or silver nitrite impregnated catalytic devices

3.12
iodine trap
device intended to trap radioiodine in gaseous radioactive effluents by using a solid sorbent in an enclosure

3.13
isotopic exchange
permutation of two isotopes of the same chemical element within a molecule

EXAMPLE An atom of iodine 131 in a CH_3I molecule in a gas form exchanges its position with a stable iodine 127 in a KI molecule impregnated on the surface of the sorbent.

3.14
nominal user flow rate
volume flow rate specified by the user which pass through the *iodine trap* (3.12) during the test

Note 1 to entry: This flow rate may be different from the flow rate specified by the manufacturer.

3.15
penetration
P
ratio of the quantity of species (particles or gas) penetrating the filter to the quantity entering it

Note 1 to entry: Penetration is always less than or equal to 1.

3.16
physical adsorption
physisorption
adsorption (3.2) with low energy of adhesion (e.g. Van der Waals) and reversible phenomenon

3.17
sorbent thickness
thickness of the sorbent layer

3.18
sorbent volume
quantity of solid sorbent present in the *iodine trap* (3.12)

3.19
specific surface area of the sorbent
total surface area (exchange surface) of the sorbent, expressed in square meter per a mass unity (in g)

Note 1 to entry: This parameter can be calculated from the surface area of a monolayer of an adsorbed probe molecule (adsorbate) at standard temperature and pressure (STP) and normalized per mass unit of sorbent.

Note 2 to entry: The specific surface area of the sorbent defines the surface available for adsorption and accessibility to adsorption sites.

Note 3 to entry: The most widely used method for determining specific surface area is the Brunauer, Emmett and Teller (BET) method. The determination of the specific surface area according to the BET method is defined in ISO 9277.

3.20
tracer gas
gas used in test

3.21
zeolite
crystalline aluminosilicate minerals that form microporous frameworks, commonly used as commercial adsorbents and catalysts

Note 1 to entry: These are commonly referred to as molecular sieves.

4 Trapping phenomena and influencing factors

4.1 Type of iodine to be filtered in nuclear facilities

Radioactive iodine is a fission product representing a serious radiological impact due to its radiotoxicity as well as its affinity toward the thyroid gland. Different isotopes of iodine (mainly ^{131}I, ^{132}I, ^{133}I, ^{135}I) can be produced from fission reactions occurring within the fuel matrix of nuclear reactors. The ^{129}I isotope can also be produced from fission process, and may be released by other nuclear facilities (e.g. fuel reprocessing plants, isotope production facilities).

Isotope ^{131}I with a half-life of about 8 days is the main contributor to iodine radiological consequences to the environment for nuclear power plants (NPPs). Isotopes with long half-lives such as ^{129}I (half-life about $1{,}6 \times 10^7$ years) could be released by spent fuel reprocessing facilities. Finally, beta minus (β−) decay iodine isotopes ^{129}I, ^{131}I, ^{132}I, ^{133}I, ^{134}I, ^{135}I and beta plus (β+) decay iodine isotopes ^{123}I, ^{124}I, ^{125}I, ^{126}I with half-lives of less than 2 months could be produced or released by laboratories or isotope production facilities.

Radioactive iodine can be released in gaseous or particle form (in most cases, iodine aerosol volatile particles could represent up to 95 % of quantity of iodine forms, but this depends on the iodine chemistry with regards to its environment inside the process or inside buildings). In this last case, these iodine aerosol particles are filtered by high efficiency particulate filter (HEPA) whose test method is not considered by this document. This document focuses on the trapping of volatile iodine compounds, represented commonly by molecular iodine (I_2) and methyl iodide (CH_3I).

4.2 Trapping phenomena

The trapping phenomena within the iodine filters are of primary importance for the in situ efficiency tests of the different radioactive products passing through the filters. For radioactive iodine, the removal efficiency results from the balance between different mechanisms for the iodine retention within the sorbent stage. These mechanisms are mainly physical, chemical phenomena, or isotopic exchanges depending on the impregnating molecules. It is worth recalling that nuclear grade activated carbons are generally co-impregnated with both potassium iodide (KI) and triethylenediamine (TEDA) molecules (e.g. 1 % mass fraction of KI and content lower than 5 % mass fraction for TEDA). The adsorption capacity of this type of activated carbons is considered in the range of about 1 g or a few grams of total iodine per kg of adsorbent (for an expected filter efficiency of about 99 %).

The main factors influencing the trapping efficiency are

a) the parameters related to the adsorbent, i.e. nature of the raw material, impregnation type and content, preparation method, granular size, bed depth, and

b) parameters specific to the gas conditioning: temperature, relative humidity, inhibitors, gas velocity described hereafter.

Some other factors influencing trapping efficiency are specific to the testing methods; these will be specified in those methods.

Three main phenomena are associated with the trapping of radioactive iodine:

— physisorption or physical adsorption;

— chemisorption or chemical adsorption;

— isotopic exchange.

NOTE Desorption can occur, because physisorption and isotopic exchange are reversible phenomena.

The relative quantification of these phenomena depends on several parameters:

— characteristics of iodine to be removed (organic, inorganic);

— nature of the adsorbent used (e.g. activated carbon, zeolite) and its characteristics (sorbent thickness, sorbent volume, specific surface area of the sorbent);

— chemical additive used to improve the performance and the stability of trapping;

— gas conditioning parameters (e.g. temperature, relative humidity).

In the following, a brief description of the main mechanisms of iodine retention is presented. Then, a small review about some influencing parameters towards the capture of iodine species is discussed.

4.3 Iodine trapping mechanisms inside porous filters

4.3.1 Physical adsorption

The physical adsorption or physisorption involves very weak interaction energy, such as Van der Waals forces. These forces are sensitive to the distance between the adsorbent and the adsorbed molecule, also known as “adsorbate”. Physisorption interaction occurs without modification of the molecular structure of the adsorbent and is totally reversible. The desorption may occur by a simple changing of gas conditioning process (temperature increase, pressure decrease, replacing the iodine flow with an inert gas…).

In addition, physisorption depends mainly on the accessibility of the adsorbate to the adsorption sites (pores). This is governed by the relative size of the adsorbate molecule to the pore size distribution of the sorbent used. Hence, this mechanism is not specific to iodine species.