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Article Sub-Title	SORGENTINA-RF
Article CopyRight	The Author(s), under exclusive licence to Società Italiana di Fisica and Springer-Verlag GmbH Germany, part of Springer Nature (This will be the copyright line in the final PDF)
Journal Name	The European Physical Journal Plus

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Schedule	Received	2 July 2021
	Revised	
	Accepted	23 October 2021

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Footnote Information	The SRF Collaboration: A list of authors and their affiliations appears at the acknowledgement section.

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Eur. Phys. J. Plus _####################_
https://doi.org/10.1140/epjp/s13360-021-02111-6

THE EUROPEAN PHYSICAL JOURNAL PLUS

Regular Article

SORGENTINA-RF project: fusion neutrons for ^{99}Mo medical radioisotope

SORGENTINA-RF

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Received: 2 July 2021 / Accepted: 23 October 2021

Abstract The SORGENTINA-RF project is presented in terms of general structure and description of the main tasks and activities to be carried out. It is devoted to the design and development of a medium power 14 MeV fusion neutron source relying on a rotating target and a deuterium/tritium ion accelerator. The main focus of the neutron facility is the production of radiopharmaceutical precursors, in particular ^{99}Mo as precursor of ^{99m}Tc, a radio-tracer used in single photon emission computed tomography. The nuclear reaction involved in the production of ^{99}Mo is the inelastic reaction ^{100}Mo(n,2n)^{99}Mo. The facility will assess the chain that starts with the irradiation of the natural molybdenum (where ^{100}Mo has an isotopic abundance of about 10%) up to the production of the so-called mother solution, a liquid solution named sodium molybdate. The facility will also make available fast and thermal neutrons beams for studies on innovative medical radioisotopes as well as materials.

1 Introduction

The development and optimization of ^{99}Mo production routes that are alternative and complementary to those presently used are strategic in the long term. ^{99}Mo is the precursor of ^{99m}Tc used as tracer in single photon emission computed tomography (SPECT), a diagnostic technique that covers more than 80% of all the nuclear medicine diagnostic procedures worldwide [1]. The gold standard for ^{99}Mo production is the irradiation of samples containing highly enriched ^{235}U with the neutrons generated at research fission reactors. At these facilities the ^{99}Mo activity obtainable in units of 6-day Ci [1] is in the range 1000–5000 6-day Ci.

In 2009, a global crisis of ^{99}Mo supply was experienced that was caused by the simultaneous and unpredicted temporary shutdown of the two main fission reactors that were

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providing a large fraction of the world demand of ^{99}Mo, i.e. HFR in Holland and the NRU in Canada [2,3]. This event prompted international organizations, such as OECD and IAEA, to ask the scientific community to propose alternative production routes for ^{99}Mo that avoid highly enriched ^{235}U (used until that period) for non-proliferation issues and fission reactors because of aging issues of the main irradiation plants.

Different routes can be thought as possible alternative to reactor-based technologies and in the following are reported representative examples, referring the reader to Refs. [4–8] for a complete overview:

1. Neutron photo-production in ^{100}Mo via the ^{100}Mo(γ,n)^{99}Mo reaction;
2. α-particle capture in ^{96}Zr via the ^{96}Zr(α,n)^{99}Mo reaction;
3. Fast proton interaction in ^{100}Mo via the ^{100}Mo(p,2n)^{99m}Tc reaction;
4. Fast neutron interaction in ^{100}Mo via the ^{100}Mo(n,2n)^{99}Mo inelastic reaction.

All these reactions make use of particle accelerators: e-LINACs in the case of the neutron photo-production, ion cyclotrons in the case of α-particles and (mainly) D-T fusion neutron sources for the inelastic reactions in ^{100}Mo.

In this context, the SORGENTINA-RF (SRF) project aims at developing a prototypical medium intensity D-T 14-MeV fusion neutron source mostly dedicated to the production of medical radioisotopes with a special focus on ^{99}Mo. Indeed, the fusion neutron route is very interesting for a series of advantages [9], but the lack of a very intense 14 MeV neutron source is a limitation to its effective use for ^{99}Mo production. SORGENTINA will be a prototype plant to assess the production route. As already pointed out in Ref. [1,10], a high power fusion neutron source may provide a Mo-99 activity in the order of a few thousands of 6-day Ci, depending on the irradiation configuration (geometry and irradiation time).

As already pointed out in Ref. [11], the development of a high-intensity fusion source needs a step-by-step approach, typical of the development of the most intense neutron sources (reactors and accelerator-driven) since the first use of neutron beams for investigations on condensed matter [12,13]. Low-intensity D-T sources are operating in different laboratories, an example being the Frascati Neutron Generator (FNG) at the ENEA's Frascati Research Centre (Italy) [14,15]. This 300 W-power compact accelerator-driven 14 MeV neutron source is the experimental starting point of the further development represented by SORGENTINA-RF, that is a remodulation of a previous conceptual project named new Sorgentina fusion source (NSFS) [16–18] devoted to fusion technology-oriented activities.

This paper is intended to provide a very general description of the project and how the main scope is thought to be reached, by implementing a series of structured and integrated design, experimental and simulation activities.

2 Scope and structure of the project

The main scope of the project is to develop a 250 kW power, accelerator-driven 14 MeV fusion neutron source that will be used to thoroughly assess the production route of ^{99}Mo from the irradiation of natural molybdenum samples up to the production of the sodium molybdate, that is the precursor of the radiopharmaceutical. Although the process was already studied and verified at a laboratory scale [10,19], the project represents a step towards a quasi-industrial plant where relevant quantities of molybdenum can be irradiated and in turn radiochemically treated in the same site. The project is performed in two main steps: first the so-called thermo-mechanical demonstrator (TMD) will be designed and made operational, while in a

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Table 1 List of the project tasks

Task# - Activity
Task 1–Civil works
Task 2–Rotating target
Task 3–Ion source
Task 4–Neutronics
Task 5–Tritium facility
Task 6–Radiochemistry facility
Task 7–Radiation protection
Task 8–Safety
Task 9–Titanium facility

second step the neutron source will be fully developed starting from the demonstrator, also comprising the radiochemical facility for molybdenum treatment.

The TMD will be composed by a rotating target and an ion accelerator that provides the 250 kW power to be evacuated by a properly designed cooling system. The accelerator, at the first stage, will operate with hydrogen ions only so to deliver power without producing neutrons.

After the design, construction and test phases of the TMD, the accelerator will operate with deuterium and tritium ions. Deuterons and tritons will be implanted onto a few microns thick titanium layer where they interact in turn producing a neutron field, the main component being that from the D-T reaction, namely $^{2}H + ^{3}H \rightarrow ^{4}He + n + 17.1$ MeV. These neutrons will be used to irradiate about 10 kg of natural molybdenum (10% natural enrichment in ^{100}Mo) to make the $^{100}Mo(n,2n)^{99}Mo$ reaction occur.

Together with the production of ^{99}Mo, the SRF plant will be used to study other medical radioisotopes as well as to implement other neutron techniques with the possible design and development of dedicated neutron extraction lines.

The SORGENTINA-RF project relies on nine tasks (listed in Table 1) where the activities related to the different aspects and components of the plant are carried out by teams composed of engineers, physicists, chemists and technicians.

2.1 Task 1–civil works

ENEA provides design, prototyping, experimental validation, safety analysis, material qualification and characterization in harsh environment, and code validation and verification supported by the main Italian universities and industries operating in the energy sector. Also, ENEA works in developing data acquisition and control systems, remote handling and maintenance.

The SORGENTINA-RF plant will be installed at the ENEA Brasimone Research Centre. Brasimone is between Florence and Bologna and it is located about 900 m above sea level: Fig. 1 shows a picture of the area. The Centre covers an area of about 4.120.000 m^2 (about 960.000 m^2 are urbanized) with several existing buildings and service infrastructures, suitable for hosting experimental halls, laboratories, storage materials and offices. Indeed, the buildings are characterized by large volumes, great heights, bridge cranes and shielding against radioactivity. The research activities carried out at Brasimone are mostly related to the technological development of fusion plants and fission reactors of the fourth generation.

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Fig. 1 Picture of the ENEA-Brasimone research centre site

Regarding the activities necessary to adapt the existing structures to accommodate the new research facilities of SORGENTINA-RF, they can be summarized as follows:

- Building works;
- Structural works;
- Mechanical and electric systems;
- General services.

The main works will be those related to the building that will host the plant. In particular: remaking of the deteriorated partial coverage, demolition of partitions, adaptation of conventional and non-conventional systems (water supply, electricity supply, gas supply), reinforcement of the attic on the ground floor, realization of the stack and the biological shield.

2.2 Task 2–the rotating target

The goal of the task is the design of the rotating target of the neutron source and of its auxiliary components (i.e. vacuum chamber and heat transfer system). The rotation of the device has a double scope:

1. Allowing the implantation of deuterium and tritium into a thin titanium layer (about 3 μm) properly deposited on the target surface;
2. Dissipation of the thermal power (250 kW) delivered by the accelerator by acting as a sort of heat pipe (Patent PCT / IB2019 / 051,972).

The main characteristics of the target's material are:

- High thermal conductivity to keep the temperature of the titanium layer below 200 °C while transferring the heat induced by the ion beam current impinging on the target;
- Low neutron activation to keep shutdown periods as short as possible, minimize the production of radioactive waste and reduce the need of remote handling systems;
- Good mechanical properties;
- Chemical compatibility with water.

Aluminium alloys were selected as the best compromise to fulfil the above criteria. The geometry of the rotating target was defined relying on computational fluid dynamic (CFD) and finite element (FEM) codes to assess: i) optimization of the thermal-hydraulic phenomena inside the heat pipe (e.g. water pumping and flow inside the rotating device); ii) estimation of the mechanical stresses inside the material. Figure 2 shows a schematic of the rotating target.

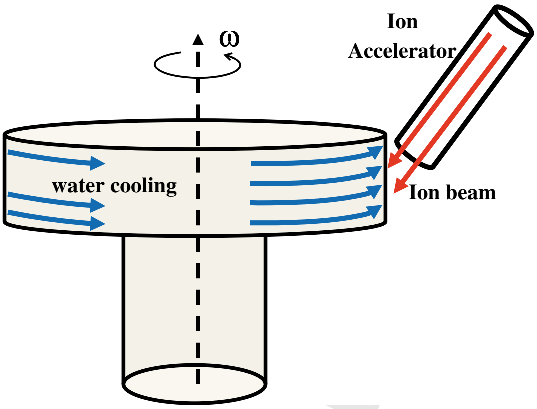

Fig. 2 Schematic of the rotating target

2.3 Task 3–the ion accelerator

The ion accelerator drives the D/T ion beam toward the rotating target where the thin titanium layer acts as a "neutron sponge". The experience gained by operating the FNG facility, the conceptual design by Martone [16,17,21] and some preliminary analytical calculations indicate that an ion beam energy of 300 keV is the most effective compromise for effective D/T implantation (typically in the order of 1.8 D/T atoms per Ti atom) and neutron production for a given ion beam current. The beam power of 250 kW at the target was fixed after evaluating a number of general aspects of the project, such as the maximum power input of the facility, the electrical consumption and the desired neutron yield for the production of a given activity of ^{99}Mo. Therefore, the nominal value of the beam current (at the target) is 833 mA (416.5 mA for each ion species). As a matter of fact, fusion reaction rate is proportional to the product of the flux of the impinging ions and the density of the target nuclei implanted in the titanium layer. In the specific case, there are two contributions to the total neutron yield rate, namely the T(D) ions of the beam impinging onto the D(T) ions implanted in the target. Both ion flux and the density of ions implanted are proportional to the current fraction of ionic species in the beam. In the reasonable assumption that the implantation rate of D and T in titanium is similar and that the D-T and T-D fusion cross sections are also similar, maximizing the neutron emission rate means maximizing the product of the ion current fractions of D and T, which brings to an ion beam made by 50% of D+ and 50% of T+ (in terms of ion current fraction). Of course together with D-T and T-D fusion neutrons, the spectrum emerging from the target presents contributions from the D-D and T-T reactions. Nevertheless, because the reaction cross sections for these fusion processes are almost two orders of magnitude lower than the D-T cross section, only the latter provides the leading term of the overall neutron emission rate. The ion source (see Fig. 3) constitutes the engine of the neutron source. It is supposed to produce D^+/T^+ mixed beam impinging onto the rotating target with precise requirements and with high reliability and efficiency. As one of the aims of this facility is to demonstrate continuous and sustainable operation, availability and maintainability of the machine are just as important. Four aspects make the realization of such device critical and peculiar:

1. It has to work on a continuous cycle and guarantee some thousands of hours of continuous functioning between two consecutive maintenance services;

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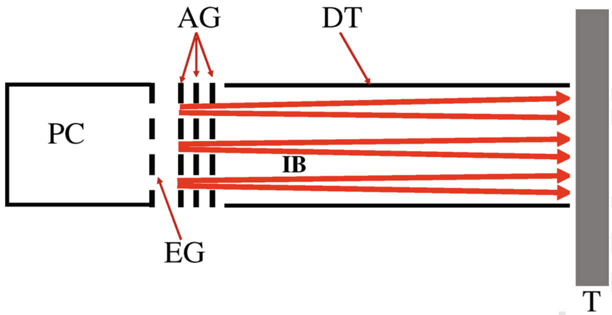

Fig. 3 Schematic of the ion source: PC is the plasma chamber, EG is the extraction grid, AG the acceleration grid, DT is the drift tube, IB is the ion beam, and T is the rotating target

2. It has to produce a mixed beam of D^+ and T^+ which composition has to be controlled and kept within certain range to preserve the neutron production;
3. It must be fed by a radioactive gas as tritium and this brings up a number of issues related to tritium-handling and contamination of some components of the ion source which must be adapted or redesigned;
4. The required beam characteristics in terms of energy and current are not typically present at the same time among positive ion sources and this increases the workload at least in the design phase, in particular the design of the acceleration stage at 300 kV.

Starting from the work of Martone [20], a size scaling with respect to the source power was performed. The definition of the beam parameters has to be established along with the optimized tritium cycle to guarantee the economically profitable production of neutrons and thus of ^{99}Mo. In this regard, a simplified time-dependent numerical model was developed to calculate, among the others:

- The amount of deuterium and of tritium implanted into the titanium layer and the one released in the vacuum chamber to be removed and recycled;
- The helium generated by fusion reactions;
- The neutron yield of the machine as some parameters are changed.

One of the first points that has been fixed is the layout of the machine, i.e. a linear device featuring a common plasma chamber (where the plasma composed by D and T ions is created) and extraction and acceleration stages optimized for both isotopes. A closed-loop system for recycling the large amount of D and T released in the vacuum chamber of the target is foreseen as well.

The tritium release rate of 8.35×10^{14} atoms mA^{-1} s^{-1} from a target of titanium under ion bombardment was taken from Ref. [20] and is in line with other experimental values found in literature [22,23]. In an ideal case (considering a gas efficiency of the ion source of 100%), about 1.56×10^{-2} mol/h of both tritium and deuterium are necessary, i.e. about 0.75 g/day of deuterium and 1.12 g/day of tritium. As the fraction of tritium that reacts to produce neutrons is 10 - 11 orders of magnitude lower (considering the expected neutron yield) than that used to feed the beam, the same amount of tritium would be released in the vacuum chamber daily. Figure 4 shows the time profile of the value (in kEur) of tritium accumulated in a single day of operation. Noteworthy, the discontinuity occurs when the target reaches the saturation of tritium implanted. Then, in order to achieve a cost-sustainable operation a D/T closed-loop is necessary. As general prescriptions for the design of the device, technologies

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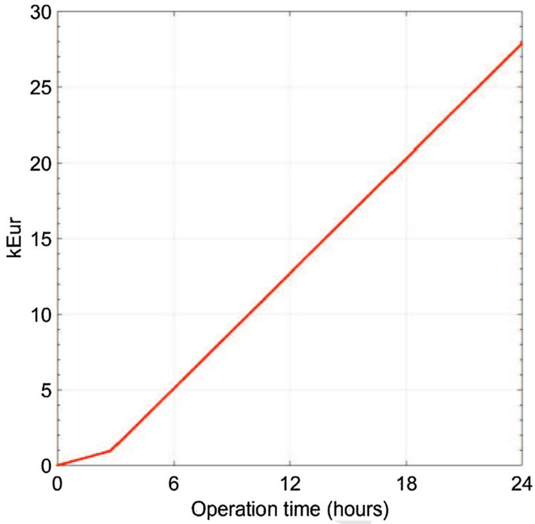

Fig. 4 Value (in kEur) of tritium accumulated in vacuum chamber in a single day without recycling

already developed for positive ion sources employed for neutral beam injectors of proven reliability have been preferred.

2.4 Task 4–neutronics

The nuclear analyses in the framework of the SORGENTINA-RF project are devoted to the design and development of the facility and involve several aspects, such as the optimization of its performances in terms of ^{99}Mo production, the assessment of the nuclear loads on its components and the design of shielding elements to mitigate the effects of neutron streaming. In particular, the following key issues have been identified and will be addressed during the SORGENTINA-RF design phase:

- Enhancement of ^{99}Mo production, achieving more than 1 Ci at the end of each irradiation session;
- Design of the bioshield that protects the environment and personnel from the radiation streaming effects;
- Assessment of the nuclear loads (neutron radiation damage, He production, nuclear heating) on the facility components to provide input for structural analyses;
- Design of the beamlines through the identification of proper pre-moderator/moderator components according to the foreseen scientific applications;
- Radwaste analysis to provide the radioactive inventory of activated materials during operations and decommissioning.

All the above-mentioned analyses are carried out by means of the MCNP (Monte Carlo N-Particle) Monte Carlo code (version 5 and 6) [24], coupled to proper nuclear data libraries (JEFF [25], FENDL [26]). As far as the evaluation of the ^{99}Mo activity and the radwaste assessment are concerned, the FISPACT-II [27] inventory code is used, complemented with the EAF-2010 [28] as activation-transmutation libraries. The MCNP SORGENTINA-RF neutron source is specifically developed on the basis of that previously implemented for FNG that has been extensively tested and benchmarked through experimental campaigns and computational simulations [29,30]. It extends over the area where the 300 keV deuteron

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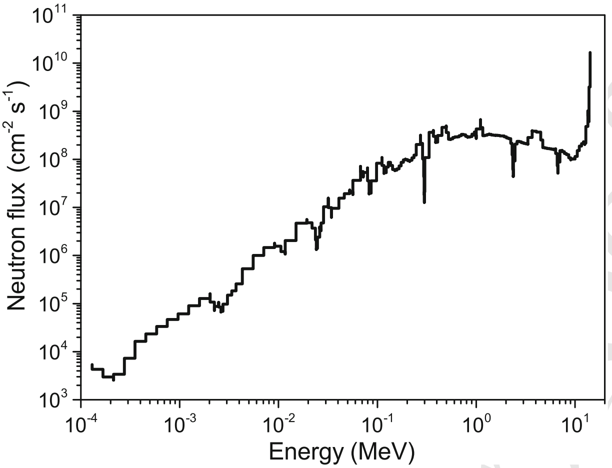

Fig. 5 Typical neutron spectrum emerging from the SORGENTINA's target due to all combinations of D and T interactions

beam impinges on the rotating target and takes into account the neutron emissivity per solid angle per energy from the D-T reactions.

The average neutron spectra impinging on the ^{100}Mo target have been preliminary assessed with MCNP for different geometrical configurations and for an expected nominal neutron emission rate in the range 5-7 $\cdot 10^{13}$ s^{-1}, allowing at predicting that more than 1 Ci of ^{99}Mo per day after 24 h of neutron irradiation is achievable. Figure 5 shows the typical spectrum provided by the overall set of fusion reactions occurring via beam-target interactions.

A proper layout for the bioshield embedding the neutron source is presently under development. The nuclear analyses require the assessment of neutron and gamma fluxes in relevant locations inside and outside the hall, along with their spatial distribution and spectra. The studies performed up to now lead to the definition of a promising structure for the bioshield, relying on ordinary/baritic concrete, and a 'dogleg' (i.e. labyrinth) access corridor to the main hall that allows a suitable protection to the entrance door (see Fig. 6). The thickness of each layer of concrete (two layers of standard and one layer of baritic) is 1 m. This configuration was designed relying on the constraint to have 10 μSv/h dose at the external face of the bioshield during operation.

The evaluation of the nuclear loads on the SORGENTINA-RF subcomponents is aimed at assessing their structural integrity and proper functionality during operations. The nuclear responses can be summarized as follows:

- Neutron and gamma flux spatial distribution;
- Nuclear heating, considering both the contribution of neutrons and secondary gamma-rays;
- Damage in terms of displacements per atoms (dpa);
- He-production in terms of atomic parts per million (appm).

The estimation of the neutron and gamma-ray flux spatial distribution provides an overview of the radiation field inside the SORGENTINA-RF neutron source hall. The nuclear heating data are used as input for thermo-mechanical analyses aimed at verifying the structural integrity of specific subcomponents, especially for the design of the rotating target cooling system. The evaluation of the damage is useful to assess the effect of radiation on sensitive elements,

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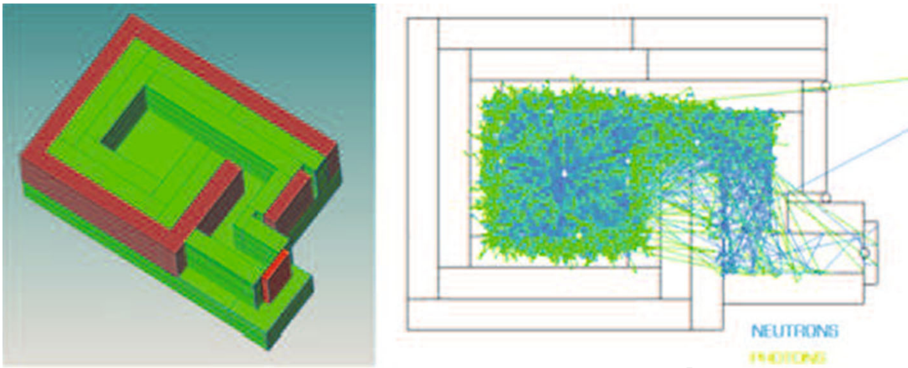

Fig. 6 (Left panel) 3D representation of the bioshielding; (right panel) visualization of neutron and γ-ray fields inside the bioshield obtained by means of the MCNP code. The roof is not depicted, and it features a total thickness of 3 m

Table 2 Neutron damage and cumulative dose for the ion source main components: SMG is the SmCo magnet, CG1/1/3 are copper grids, and AI-1/2 are the alumina inserts

Parameter	SCM	CG-1	CG-2	CG-3	AI-1	AI-2
Damage (dpa/FPY)	1.0E-02	6.7E-03	6.2E-03	5.2E-03	1.7E-03	1.9E-03
Cumulative dose (Gy/y)	5.3E+06	1.4E+06	3.3E+06	1.1E+06	8.8E+05	1.0E+06

such as grids, insulators and magnet of the ion source, in order to prevent the degradation of the physical properties of the materials. Table 2 provides a preliminary assessment of the damage (dpa/FPY, FPY meaning Full Power Year) and integrated dose (Gy/y) on specific components, such as SmCo magnet, copper grids and alumina inserts, for an ion source equipped with a 1 m drift tube (see Fig. 7). Moreover, the radiation field expected around the Sorgentina neutron source is shown in Fig. 7 as a 3D maps of the neutron flux density.

Beyond the main scope of ^{99}Mo production, the neutrons generated during the SORGENTINA-RF experimental campaigns represent a remarkable source that can be exploited for a wide range of scientific and industry-relevant applications. Extraction lines for 14 MeV and thermalized neutron beams are under consideration, and some preliminary studies on moderator systems effective for almost monochromatic fusion neutron fields have been conducted at FNG [31–34].

The assessment of the expected dose inside the bioshield due to materials activation is fundamental to determine a maintenance schedule for ordinary and/or extraordinary interventions. Consequently, all elements and equipment installed in the machine must undergo an evaluation of the radioactive waste produced during operations and still present at the decommissioning of the plant. It is worth to stress that the driving criteria for the choice of the materials for the structural components of the machine is based on the minimization of the activation (e.g. usage of 316L(N)-IG austenitic stainless steel and aluminium alloys). A complete activation analysis for the SORGENTINA-RF components is foreseen using the FISPACT-II inventory code, according to a reliable irradiation scenario. The neutron spectra (vitamin J energy group structure, 175 energy bins), provided as input for the activation calculations, will be evaluated in specific positions using the MCNP code. For each subcom-

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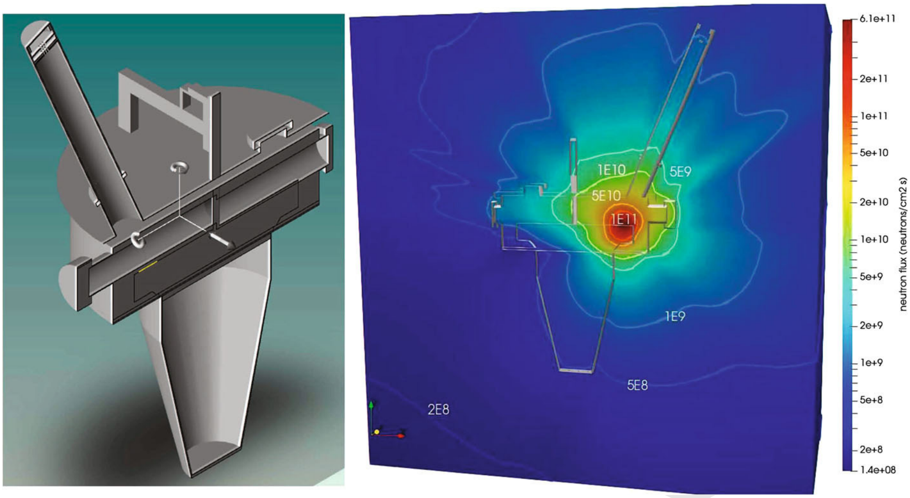

Fig. 7 (Left) 3D view of the Sorgentina MCNP model, with an enlargement of the ion source main components.; (right) 3D maps of the neutron flux density (units are cm^{-2} s^{-1})

ponent, relevant data for the classification of radioactive waste (e.g. the total activity, surface effective dose rate, dose rate at 1 m distance, tritium content and tritium activity) will be provided.

2.5 Task 5–the tritium facility

The main scope of the tritium facility is to process and recycle deuterium and tritium used in the vacuum chamber for the high-energy neutrons production. The ion beam, which is the source of both deuterium and tritium, is fuelled by a pure D-T gas mixture. Therefore, the other impurities from the vacuum chamber as titanium, helium, argon, oxygen, etc., have to be removed from the stripping gas before to route the Q_2 (Q=D,T) isotopes to the fuel management system. It should be pointed out that particulates such as TiD and TiT have to be removed from the exhaust gas stream by appropriate filters characterized by high efficiency. The tritium system has to be placed inside a dedicated glove box. The system shall be able to handle the exhaust gas with an impurity composition of about 1 at. %, whereas the concentration of H in the purified gas should be less than 2%. A schematic representation of the process is reported in Fig. 8. The system shall be able to handle the exhaust gas flow rate of about 0.036 mol/h D-T from vacuum chamber; the other main operative conditions are reported in Table 3. In Fig. 9, the process flow diagram (PFD) of the tritium facility is reported.

Four main subsystems can be identified, namely:

- The vacuum system (100);
- The tritium control system (200);
- The Pd/Ag permeator system (300);
- The tritium getter storage system (400).

The vacuum system operates at a pressure of 10^{-3} Pa and has the function to pump the D-T mixture coming from the vacuum chamber. The filters, needed to remove impurities from the

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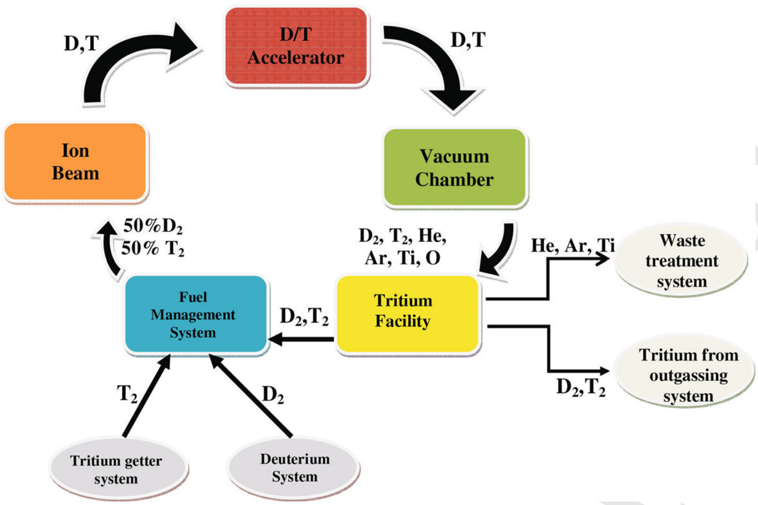

Fig. 8 Schematic process of SRF tritium system

Table 3 Operative conditions of tritium facility

Parameter	Value	Units
Q_2 pres. in vacuum chamber	$1 \cdot 10^{-3}$	(Pa)
Q_2 pres. in plasma chamber	0.2–0.3	(Pa)
Q_2 pres. in tritium facility	$1 \cdot 10^{2}$	(Pa)
D-T pumping flow rate	0.036	(mol/h)
Gas mixture	$He/T_2/D_2$	–
Impurities	Ti, O_2, Ar	–

gas stream, are redundant in order to guarantee a continue operation. Vacuum is generated by means of a turbo-molecular pump backed by a dry scroll pump or rotary vane pump.

The tritium control system is designed to allow a dynamic control of the deuterium and tritium inventories; the D-T mixture coming from the vacuum chamber is pumped into a pressurized storage tank (V1) where it is accumulated prior to be sent to the permeator system. According to the accelerator needs, two alternatives storage tanks (V2, V3) can be used to compensate the D-T flow in order to keep the concentration in its stoichiometric ratio. The gas composition is evaluated by a dedicated mass spectrometer and a ionization chamber. Figure 9 shows a schematic diagram of the tritium system envisaged for the plant.

Tritium is stored by a getter system, which is one of the most convenient ways of handling tritium [35]. Several metallic materials can be used for this purpose, for example uranium, palladium, zirconium, titanium or lanthanum–nickel alloys. The getter bed is provided with a dedicated heating system which allows the solubilized tritium to be desorbed and with a control valve which opens once the desired pressure is reached. On the other hand, deuterium is stored within a cylinder provided by a pressure reducer. The different subsystems are thermally traced in order to remove water vapour and other impurities from the stainless steel pipings and components before the facility operation or during the maintenance period. Finally, the technology adopted to remove tritium and deuterium from the exhaust gas coming from the vacuum chamber is the Pd/Ag membrane reactor [36], for which a scheme of the

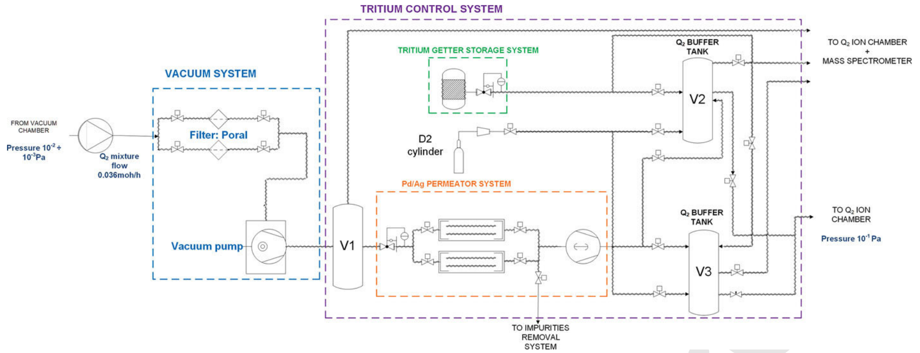

Fig. 9 PFD of tritium facility connected to the vacuum chamber and the ion chamber. Tritium system is constituted by vacuum system, tritium control monitor integrated within two glove box, one dedicated to tritium getter storage system and one related Pd/Ag permeator system and auxiliary systems for pressure control management. Gas composition is monitored by means of ion chamber and mass spectrometer

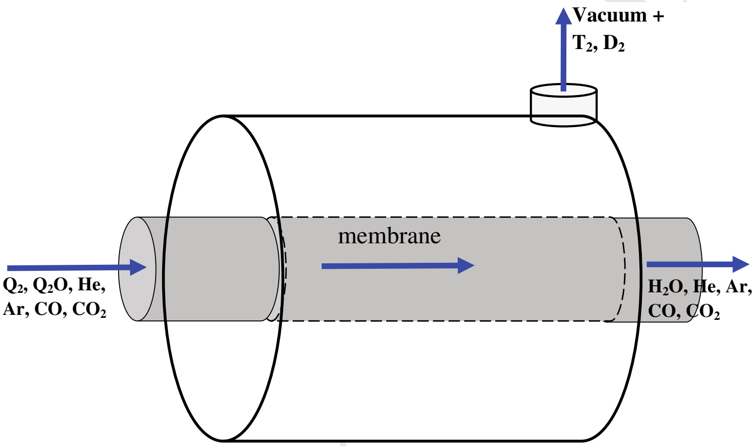

Fig. 10 Functioning scheme of a Pd/Ag membrane reactor

process is illustrated in Fig. 10. This technology is based on the phenomenon of tritium permeation through a membrane towards a secondary side where vacuum is present. In this way, a pressure gradient is established promoting tritium extraction and catalyst particles promote T_2O and D_2O separation; it has to be observed that these chemical species could be formed due to the presence of oxygen in the system. The impurities removed are then sent to the impurities removal system.

2.6 Task 6–the radiochemistry facility

The activity of this task is the design, development and implementation of the chemical processes by means of due components for the production of the sodium molybdate Na_2MoO_4, i.e. the so-called mother solution from which ^{99m}Tc, in the form of pertechnetate, is obtained for biodiagnostic and medical imaging purposes. To achieve this aim, the chemical processes involved in the dissolution of the irradiated target must be modelled and experimentally assessed on a laboratory scale and, once these have been established, the necessary systems

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must be designed and built to allow large-scale and continuous production. This molybdate solution must be prepared as quickly as possible in order to avoid that the specific activity of the target decreases too much.

Schematizing the process in the SORGENTINA-RF plant, the irradiated molybdenum target is transferred into shielded hot cells where the chemical processes take place. The high activity of the target requires the use of appropriate hot cells and the development of almost fully automated processes to protect the operators. From a chemical point of view, it can be assumed that the irradiation has a reduced impact on the process; nevertheless, it might be possible that the reactivity can be modified as the irradiation process may induce crystallographic and structural defects, which the presence may even be beneficial in increasing the speed of the reactions. This is an aspect that requires investigations within the activity of the task.

Molybdenum belongs to the chromium group and has a rich chemistry because it has oxidation states ranging from -II to +VI and coordination numbers from 0 to 8 [37]. Higher oxidation states are more relevant and in general compounds of Mo(VI) are more soluble, so that the goal of the dissolution process is to form a solution containing $MoO_4{}^{2-}$ ions. This metal has several analogies with the W chemistry, but it reacts more easily with (strong) inorganic acids and oxidants such as hydrogen peroxide.

This feature can be exploited to use a chemical dissolution method that can be considered "green". In fact, it is possible to avoid the use of strong acids which can be very polluting and resort to the reaction with hydrogen peroxide, whose waste products are only oxygen and water vapour. The most effective conditions for this process in terms of temperature, pressure and concentration to achieve complete transformation in the expected time will be determined.

Another activity is related to the quality control: quality and impurities assessment of the material before irradiation must be checked, as well as fulfilment of the Pharmacopoeia requirements of the mother solution. This can be done by mass spectroscopy with a quadruple spectrometer. In general, the production method of ^{99}Mo and consequently of ^{99m}Tc proposed in the SORGENTINA-RF project has, beyond the technical difficulties to be overcome, some great advantages compared to more traditional methods. The first is that the yield is potentially higher in terms of the ratio between the mass of the material produced and that of the initial material and therefore the quantity of waste products is much less. Furthermore, the quality of the product can be higher thanks to the lower concentration of impurities and secondary phases. Finally, greater versatility of the system can be imagined. The conversion of the plant to the production of other radioisotopes, useful in medicine, such as ^{68}Ga or ^{64}Cu, can be designed and performed with relative ease, whereas a traditional implant would require large reconstructions. Figure 11 shows a schematic of the radiochemistry facility.

2.7 Task 7–radiation protection

A preventive safety analysis of SORGENTINA-RF and the related activities has been carried out in order to identify ways in which potential exposures could occur. The sources of ionizing radiation present in the facility are direct radiation, prompt and delayed gamma radiation due to material activation, tritium diffusion, environmental contamination in detail [38]:

– The primary neutronic field resulting from the fusion reactions;
– The gamma radiation generated from neutrons interaction with the machine components and the shielding;
– The gamma radiation emitted by activated products in the machine components and in the shielding;

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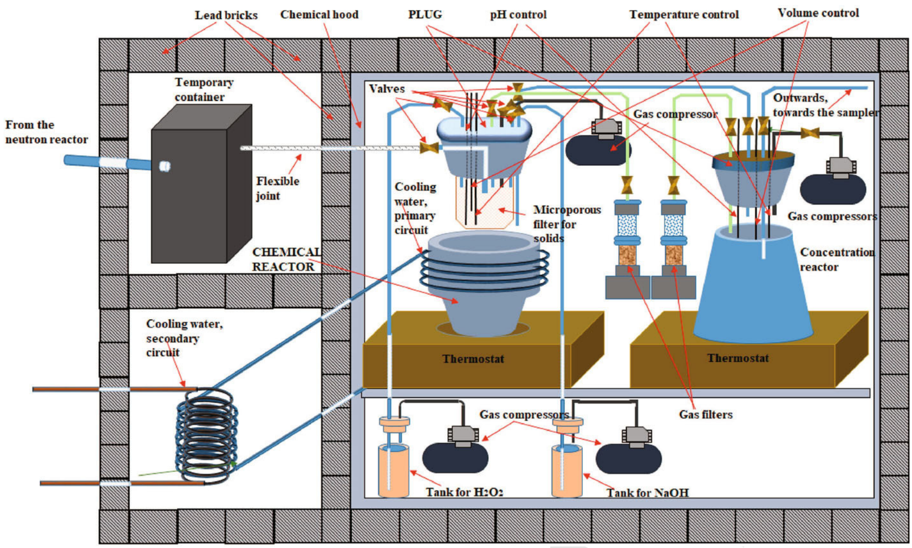

Fig. 11 Schematic representation of the radiochemistry facility

- Loose contamination from activated dust (mainly due to the activation of Ti) generated in the machine components;
- Activated corrosion products (ACPs) generated in the cooling loops after the activation of the pipes inner surface and of the corrosion products in the cooling fluid that reach high neutron flux regions of the circuit;
- Activated cooling water;
- Activated (mainly ^{41}Ar);
- Tritium used as fuel for the fusion reaction or produced in the fusion reactions;
- Radioactive waste;
- Unsealed radioactive substances handled in the radiochemistry facility;
- Liquid and gaseous effluents of the radiochemistry facility.

These source terms can expose workers and population to a risk of both external exposure and internal contamination. In the design phase an accurate qualitative and quantitative characterization of the source terms is conducted, and various technical and organizational requirements are addressed relating to each source term, including [39]:

- Correct spatial arrangement and organization of premises;
- Appropriate ventilation system;
- Installation of special equipment (such as the biological shielding around the neutron source);
- Adequate solutions for the management and storage of solid and liquid waste and gaseous and liquid effluents;
- Working procedures aimed to the safe management of activities involving risks of exposure to ionizing radiation;
- Monitoring programs and systems.

Consequently, the choice of the building that will host SORGENTINA-RF has been made also according to radiation protection criteria. The radiochemistry facility is next to the biological shielding that contains the accelerator structure in order to obtain the shortest

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possible transport route for the irradiated materials that have to be processed. An adequate number of air changes per hour and pressure gradients will be determined depending on the usage, dust contamination, estimated air activation and tritium concentration in the air, according to national and international guidelines [39]. Whenever possible, all structural materials of the machine will be reduced-activation materials for fusion, such as reduced activation steels and non-ferrous low-activation alloys, in order to reduce the exposure of personnel and the production of radioactive waste. To this respect, some radioactive waste is expected to be produced, even if machines that use nuclear fusion reactions do not present critical points [40]. As far as solid waste is concerned, a volume-reduction strategy based on appropriate material characterization will be considered for identifying both those materials that can be safely released to a conventional landfill and materials that have to be disposed and managed as proper radioactive waste. Liquid effluents potentially produced will be managed according to national regulations and international guidelines [41,42]. Also the impact on the population of the gaseous releases during operation will be estimated, which in general is not relevant [43]. In the planning of operations, a prior assessment of individual doses and risks of workers has been done. A monitoring program will be established for each pathway of exposure, specifying media to be sampled, location and frequency of sampling and measurements, radionuclides to be quantified and monitoring systems to be used [44]. In order to operate according to the current Italian legislation [45], Sorgentina-RF needs a Category A license since it will feature a neutron emission rate higher than 10^7 s^{-1} all over the solid angle averaged over time (in 1 year of operation). The request of licensing has to be accompanied by a detailed technical report related to the radiological protection of workers and population, including at least:

– Description of the installation and the activities carried out;
– Suitability of the chosen site, buildings, and structures;
– Radiation protection structures and organization, such as classification of areas and personnel;
– Shielding calculation;
– Dose rate assessment;
– Accident analysis and relevant consequences;
– Radioactive waste assessment and management;
– Dose constraints for the process of optimization.

The main technical issues are related to the level of protection guaranteed to workers and to the public during normal and accident situations. The critical aspect about population safety is related to the release of radioactive waste into the environment. A thorough analysis of scenarios involving potential exposures, considering both the neutron source and the radiochemistry facility, has been conducted, also estimating the vulnerability of the activities carried out in the facility to extreme weather events [46]. During operation the physical surveillance of Radiation Protection will be ensured according to the national law [45]. As an integral part of programs for source monitoring, environmental monitoring and individual monitoring, a quality assurance program will be provided to verify that each task has met its objectives and that any necessary corrective actions have been implemented, through quality control mechanisms and procedures such as internal audits [47].

2.8 Task 8–safety

As required for any advanced nuclear system, a throughout safety assessment has been scheduled for SORGENTINA-RF to identify any hazard related to the operation of the facility,

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possibly leading to accident events under any of the foreseen plant states. The safety assessment process integrates the complementary techniques [48] of Probabilistic and Deterministic Safety Analyses (PSA and DSA). Both demand consideration of Postulated Initiating Events (PIEs), namely possible accident initiators which are classified according to the representativeness and the severity of their consequences.

The PSA aims mainly at the identification of the PIEs. For SORGENTINA-RF a Functional Failure Mode and Effect Analysis (FFMEA) [49] methodology is exploited to define possible accident initiators. Starting from the whole functional analysis model of the system, a top-down approach is followed. The FFMEA tables will include:

- Function identification and classification (e.g. process, safety and system protection);
- Equipment and components for each function;
- Function failure modes;
- Possible causes for the loss of function associated to a specific failure mode;
- Possible consequences in terms of machine damage, radioactive inventory mobilization through the different containment barriers, dose to workers and population:
- Means to prevent the causes or mitigate the consequences of failure;
- Identification of the representative PIEs for a single elementary failure.

DSA is performed by means of validated simulation codes to investigate the accidental sequences that results from PIEs [48]. Compliance between the results obtained and the radiological acceptance criteria must be verified. From this perspective, source terms and plant hazards have been characterized to trace the amount and isotopic composition of the material postulated to be released from SORGENTINA-RF. These source terms include:

- Tritium, from its dedicated facility, the target and the accelerator;
- Activated materials such as solid dusts, gas (^{41}Ar or nitrogen), liquids and corrosion products;
- Thermal and chemical energies which might be released in accidental sequences damaging confinement barriers.

In SORGENTINA-RF, tritium released in the vacuum chamber hosting the rotating target will be recovered by the tritium facility through penetrations in the bioshield. An essential part of the safety analyses is dedicated to avoid any criticality of the recycling process which might release tritium in normal operation or accidental conditions. Confinement and containment performances of the vacuum system, the primary cooling circuit and the bioshield will be addressed. Water activation and the transport of contaminants in the secondary heat transfer loop will be taken into account to assess if they might be considered as radiological hazard for workers and population.

A closing activity of occupational safety is envisaged to support the radiation protection team to estimate the collective dose for workers employed in SORGENTINA-RF operation. Due to the uniqueness of the system, specific maintenance plans must be developed in accordance with the ALARA principle [50]. To estimate properly occupational collective effective doses for preventive and corrective maintenance operations, dose rates must be combined with maintenance data in terms of type of intervention, number of operators involved, yearly frequency, elementary work effort (i.e. execution time), ancillary safety equipment required (e.g. masks, suits and gloves) that might impede agile movement.

2.9 Task 9–the titanium facility

The so-called titanium facility is needed to recover the titanium atoms sputtered by deuterium and tritium ions to maintain constant the thickness of the titanium layer. In Fig. 12, two

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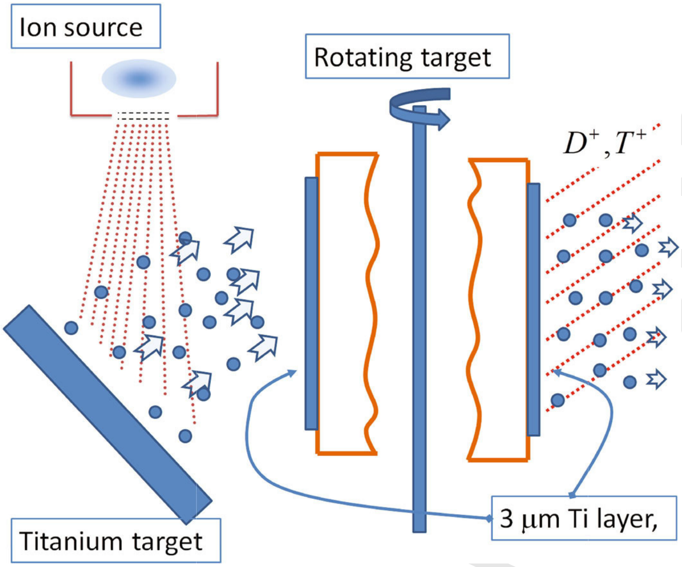

Fig. 12 Layout of how the titanium facility works. The ion source (top left) delivers Ar ions on the Ti sputtering target

different sides of the rotating target are displayed where the D/T ion beam sputters the Ti layer (right) and where the layer is recovered at the initial thickness of (3 μm) by collecting the atoms coming from the target of Ti (left). The aim of the titanium facility is to restore the Ti layers, consumed by the action of the D-T ion beam, to the initial value. This is provided by means of a sputtering-based Ti deposition relying on an ion source delivering Ar ions on a Ti target (see top left in Fig. 12). The parameter which governs the rate of erosion of a bombarded material is the sputtering yield that depends on the ion energy, the density of the ions hitting the surface and the relative mass of ions and target atoms. In order to estimate the sputtering yield of the titanium layer bombarded by deuterium and tritium ions of 300 keV energy, preliminary simulations have been performed by a Monte Carlo code, namely "The Stopping and Range of Ions in Matter" (SRIM) [51]. Depending on the properties of the ion source, a 300 keV tritium and deuterium beam may have a different composition in terms of D^+, D_2^+, D_3^+,T^+, T_2^+ and T_3^+ [52–54]. Considering the same fraction of the species composition for deuterium and tritium ions, $\delta_{D^+,T^+} = 0.74$, $\delta_{D_2^+,T_2^+} = 0.21$ and $\delta_{D_3^+,T_3^+} = 0.05$ [54], the erosion rate of titanium is 0.22 μm per day. Thus, the titanium thickness should be approximately restored each two days as a consequence of the maximum penetration depth, 2.1 μm, of the deuterium and tritium ions.

3 Conclusions and perspectives

A brief and general overview of the SORGENTINA-RF project was presented. The project is devoted to the design and realization of an accelerator-driven fusion neutron source featuring a power of 250 kW and an expected neutron emission rate in the order of 5–7 × 10^{13} s^{-1}. The plant is mostly devoted to production of medical radioisotopes with an initial main stream activity on ^{99}Mo production. Also, neutron extraction lines are under consideration to use both

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fast and thermal neutrons for applications in material science. The plant relies on a rotating target and a D/T ion accelerator to produce fusion neutrons and a series of operational and ancillary components. The main activities of the project have been highlighted to describe the lines of research and development that will be implemented during the different phases identified.

Acknowledgements The authors acknowledge the Regione Emilia Romagna-ENEA agreement for the project "SORGENTINA RF–Thermomechanical Demonstration".

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Data Availability Statement The datasets generated during and/or analysed during the current study are not publicly available due patent but are available from the corresponding author on reasonable request.

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