STATUS OF CEA SPALLATION MODULES FOR ADS

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ABSTRACT

In the framework of CEA studies on ADS dedicated to waste transmutation, a liquid metal reference concept and an alternative solid target have been evaluated to produce neutrons inside the *spallation module*. This work examines the design (neutronic, thermohydraulic and mechanical aspects) and the performances of both options. It is shown that a liquid PbBi target offers more possibilities regarding to high protons current densities (possible industrial extrapolation) but that a solid target made with tungsten particles offers also interesting ability to create a neutrons flux appropriated (strong spectrum and flat axial distribution) to an sub-critical core dedicated to incineration.

Keywords: ADS, transmutation, spallation, target, wastes.

1- INTRODUCTION

In the framework of CEA studies on hybrid systems dedicated to waste transmutation, one of the most innovating tasks remains the design of the spallation target which produces the neutrons required to maintain the fission reaction in the surrounding sub-critical core. It has appeared interesting to think this target as a compact and, as far as possible, independent module, called *spallation module*.

In the goal of an industrial plant dedicated to waste incineration, a liquid metal reference design and a solid tungsten alternative concept of a spallation module have been proposed and evaluated. The present study describes these designs and the operating conditions of a spallation module for a typical industrial plant with a focus on the main technical options: beam window, flow configuration, choice of materials, etc. Design studies have included a wide variety of assessments like mechanical and thermal-hydraulic analyses, irradiation damages in the materials, corrosion and lifetime evaluation. Solid and liquid targets have been evaluated in order to examine advantages and drawbacks of both concepts.

2- THE STATE OF THE ART ON SPALLATION TARGETS

2.1 Introduction

Whether they are solid or liquid, interesting materials for spallation reactions are those who have a high density. Indeed, the excess of neutrons with regard to the number of protons in the nucleus ensures that many neutrons will be ejected after the collision. Moreover, the macroscopic spallation cross-section is proportional to the volumic number of atoms, which assures that the protons beam will be efficiently used.

2.2 State of the art in view of ADS applications

Up to now, all targets have been built with solid spallation materials. The main reasons are that:

- Most of heavy materials are solid at usual temperatures.
- These materials are often refractory metals which allow important heat deposition.
- Spallation products can be enclosed inside.
- The target material can be used as structure.

However, to make intense neutrons sources, the power evacuation in the target can limit its performances:

- The coolant volume reduces the density of the spallation material.
- This coolant sustains an important neutrons and protons flux.
- Irradiation damages (dpa, gas production...) shorten the target lifetime.

Among usual materials, we have zirconium (SINQ), depleted uranium (ISIS), tantalum (ISIS), tungsten (APT)... They are used under the shapes of water cooled plates or ladder. Considering this limitation in the power evacuation, in the ADS development studies, the search for intense neutrons sources has induced, for future instruments of diffraction measures and for irradiation tools (ESS, Megapie, Myrrha...) to consider liquid spallation materials. The advantages of these targets are:

- The spallation material is its own coolant (compatibility with the neutrons and protons fluxes, important heat transfer coefficients, high compactness: no void fraction).

- The target is part of a loop (spallation products are spread over a bigger volume, the material can be withdrawn by draining before maintenance, a minimum cooling can be assured by natural convection in case of primary pumps failure).
- The target material faces no structural damages (but the problem is reported on the window and the containment vessel).

However, some drawbacks remain:

- Just a few heavy metals are liquid at common temperatures, for instance: mercury (T_{fus}=-40°C) and lead-bismuth eutectic (T_{fus}=127°C).
- In the case of lead-bismuth eutectic, an ancillary heater is necessary to keep the liquid state during the accelerator shutdown and the startup phases. Moreover, states changes can entail important mechanical stresses on the whole loop.
- The target vessel faces corrosion with the liquid metal, irradiation or even shock waves in case of pulse working.
- The whole primary system contains an highly activated fluid so that the installation becomes complex.
- The window, which is already stressed by the proton beam, is directly in contact with the liquid target. Besides, a concept without window (Myrrha at SCK/CEN) is studied.

Because of the high level of deposited power (up to several kW/cm^3) appearing in the ADS applications, and considering the exposed advantages and drawbacks of both concepts, CEA has chosen the liquid metal option for its reference spallation module. The next section examines the performances of the latter and of an alternative concept.

3- CEA SPALLATION MODULES FOR ADS APPLICATIONS

3.1 Function

On the basis of a functional analysis, it appears that the function of the spallation module (cf. Fig. 1) is before all to give neutrons to the sub-critical core with respect to the vacuum of the protons accelerator, whatever the working conditions of the reactor. It has also to evacuate the power deposited by the spallation reactions and to reduce to the minimum the irradiation of the accelerator structures. Finally, this module takes part also in the installation safety and in the protection of the accelerator by isolating the spallation products inside the target, in accidental situation.

This module consists in a beam window, a spallation material, which can be solid or liquid, internals for fluid guidance, a containment vessel and external ancillary equipments.

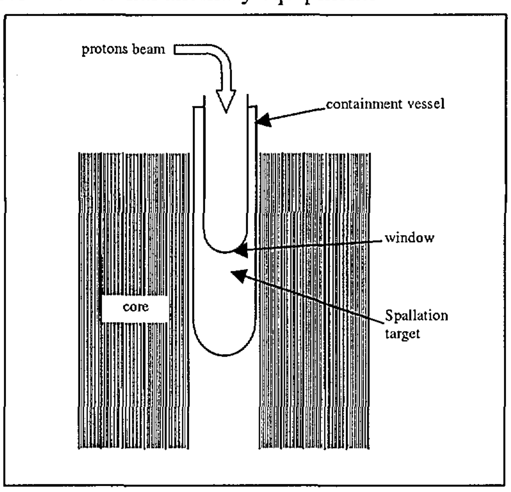

Figure 1: the spallation module in the middle of the core.

3.2 The liquid metal CEA reference spallation module

For this concept, the eutectic PbBi has been chosen due to its spallation performances, its low fusion temperature (T_{fus}=127°C) and its compatibility with martensitic steel of the target parts. Inside the module, the fluid follows a forced convection flow to assure an efficient power evacuation. The circulation direction mentioned on Fig. 2 is arbitrary, and will be discussed below. A grid is set up below the window to create a turbulent flow, efficient enough to improve the cooling conditions. The inside pressure has been set to the conservative value of 3 bar on the top of the module (about 10 bar at the bottom, on the window).

The structures components of the module and especially the guide tube and the window, are made of T91 steel (9% Cr), which has good thermal and mechanical properties and a good compatibility with PbBi below 550°C. The window is hemispheric, even if other profiles have been studied and their influences on cooling. Buckling of the guide tubes and the window head have been studied with the finite elements code *CASTEM* modelisations. It has been shown [1], according to design criteria [2] that for a radius of 16 cm, a 5 m high guide tube has to be at least 6 mm thick in the vertical part and that 1.5 mm was thick enough in the window part. As a result, in the study, thickness as been set to 8 mm vertically and to 1.5 mm at the head.

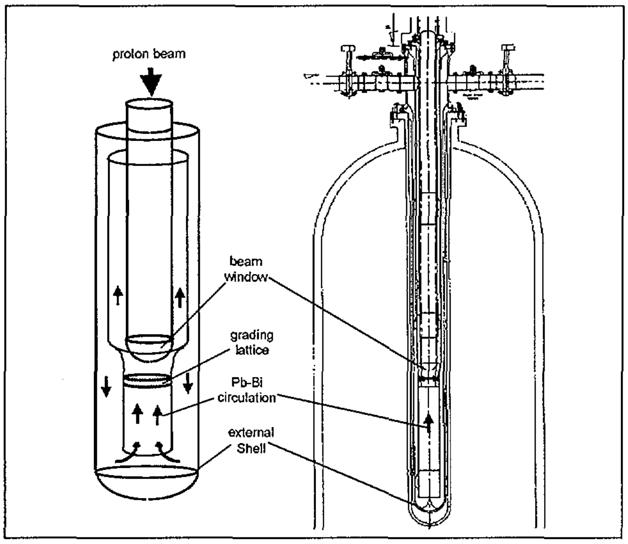

Figure 2: The CEA lead-bismuth liquid target.

In the neutronic study, made with the CEA *SPARTE* code system, it has searched an optimisation of the different factors of the system. So, a parametric study has been carried out. The protons energy and the impacted window diameter were the main variables. The former is directly related to the prospected accelerator performances and the latter is related to the spallation module size, which will have to be as small as possible. In this study, the protons beam diameter varies from 10 to 30 cm and the protons energy varies from 400 MeV to 1200 MeV. In this range, the spallation reactions produce between 7 and 38 neutrons/incident proton. This ratio increases with the beam diameter and, of course, the energy of protons. On the contrary the spectrum of the neutrons leaving the module does not depend on the beam diameter or the energy of protons. 90% of these neutrons leave the module with an energy between 100 keV and 2.5 MeV. In any case, more than 90% of the produced neutrons leave the spallation module. As a result, it appears that inside an ADS, this liquid metal spallation target has an efficiency and a strong spectrum well adapted to the transmutation of nuclear wastes.

Thanks to Fig. 3, it can be noticed that with a constant current density, the deposited energy in the impacted zone of the window (the most critical one), increases only 20% while the energy of protons goes from 400 to 1200 MeV. On the contrary, dpa (displacement per atom) depends on the energy of protons and on the beam diameter.

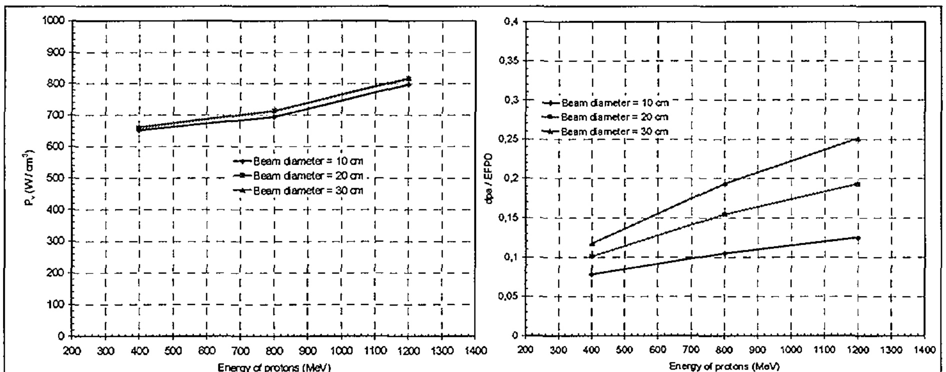

Figure 3: Heat deposition and dpa/EFPD (Equivalent Full Power Day) in the impacted zone of the window with a constant current density of 32 $\mu A/cm^2$.

In the range of energy and size of the studied proton beam, heat deposition in the liquid metal has also been examined: it shows that it can reach almost 4 kW/cm^3 (for a current density of 127 μA/cm^2). At 1 cm of the beam limit, heat deposition never exceeds 100 W/cm^3 whatever the depth. After 60 cm below the window, it never exceeds 160 W/cm^3. So, the zone where the spallation reactions take place is not extent around the beam axis.

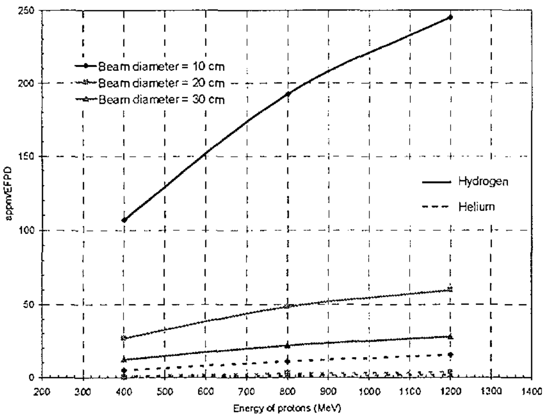

Fig. 4 shows that hydrogen is produced about 20 times more than helium. Hydrogen will diffuse in steel and be responsible of embrittlement while helium will entail swelling of the window. Between 400 and 1200 MeV, the former increases 15% and the latter increases less than 40 %. As a result, gas production depends mainly on the current density, which is the key parameter.

Figure 4: Gas production/EFPD (Equivalent Full Power Day) in the impacted zone of the window with a constant intensity of 10 mA.

The thermohydraulic study [1] of the spallation module has used the results given by the neutronic study for a beam of 800 MeV protons and a current density of 32 μA/cm^2. It has examined ascending and descending flows, and different profiles of the window (cf. Fig. 5).

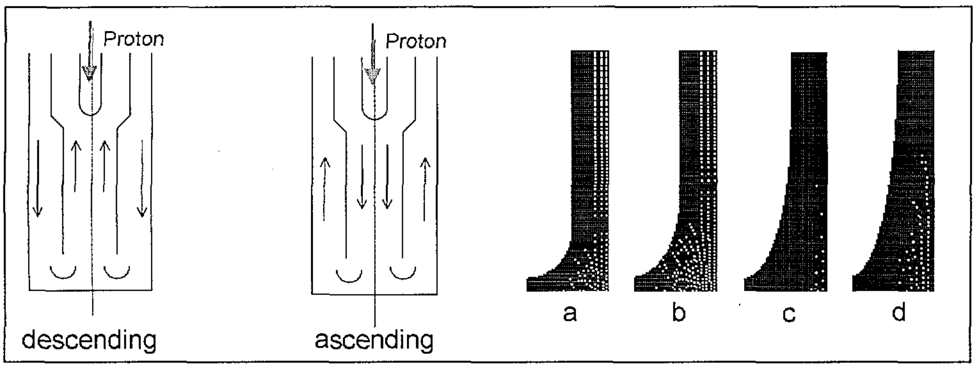

Figure 5: Types of PbBi flows inside the spallation module and types of window profiles (a=hemispheric).

A 2D calculation made with the *TRIO-U* code has shown that the PbBi temperature between its entrance in the module and the head of the window increases of about 1400°C in the case of the ascending flow, which is not acceptable regarding to the steel mechanical resistance. So, the descending flow has been kept.

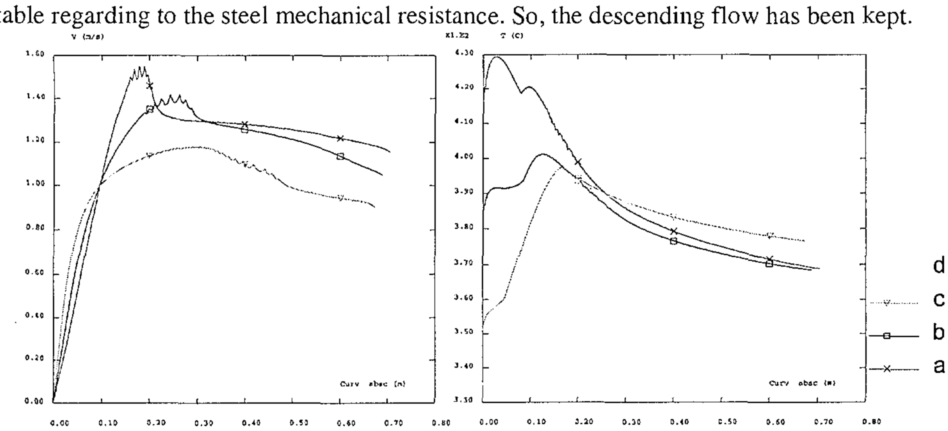

Figure 6: Velocity and temperature along the window for the different profiles, for Ve=1m/s and Te=175°C.

Similar *CASTEM-FLUIDE* calculations have shown that in the case of a descending flow, an hemispherical profile is not the best one (cf. Fig. 6) because of the velocity of the fluid which decreases with the window eccentricity: for an entrance temperature (Te) of 175°C and an entrance velocity (Ve) of 1m/s, the fluid velocity on the wall does not exceed 1.2m/s for profiles c and d. Moreover, the highest temperature in these cases is 35°C lower than in the hemispheric case. As a result, regarding to corrosion and mechanical resistance, the elliptical profiles will have more interests than the hemispherical one.

The mechanical assessment of the module has been concentrated on the beam window as it is the structural component which encounters the most severe loading. The window will indeed be submitted to both mechanical pressure and thermal loading: the pressure loading on the window is driven by the hydrostatic PbBi pressure which is supposed to be constant in the present study and has been fixed to a conservative value of about 10 bar at the bottom of the window. The thermal loading is driven by the protons current density; the nominal value for an industrial plant is not known at this stage as it is function of the current I and of the beam radius Rb. In this section, a first evaluation has been performed considering typical values I = 30 mA and Rb = 13 cm, leading to 56.5 $\mu A/cm^2$. Mechanical stress levels in the window have been evaluated with the help of the finite elements *CASTEM* code and analysed with regard to classical mechanical design criteria [2] related to risks like excessive immediate deformation, overall progressive deformation, creep rupture and corrosion. It is clear that the mechanical behaviour of the window will strongly depend on its cooling conditions. A parametric approach has then been considered aiming at defining an acceptable domain for the thermal-hydraulic cooling parameters of the window: the temperature of the liquid metal along the window (T_{PbBi}) and the convective heat transfer coefficient along the window (h).

Fig. 7 synthesises the minimum and maximum limits on T_{PbBi} as a function of h, which guarantee the respect of the design criteria. Each curve corresponds to a specific design criteria. As a preliminary general comment, it can be noted that maximum liquid metal temperatures are often imposed by mechanical criteria, the reason being to keep sufficient mechanical resistance and to limit the thermal loading. Minimum temperatures are essentially imposed by the need to keep a minimum liquid metal temperature in order to avoid solidification. A specific limitation appears also on h and corresponds to a limitation on velocity due to corrosion effects. The conjunction of all these curves delimits an acceptable domain for the cooling parameters.

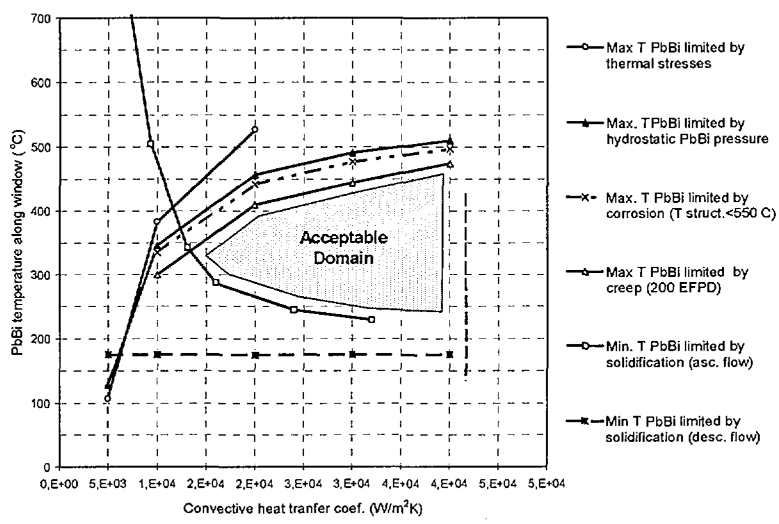

Figure 7: Acceptable operating range for the thermal-hydraulic parameters (T_{PbBi}, h) defining the cooling conditions along the window (Ep = 1.2 GeV, I = 30 mA, Rb = 13 cm, PbBi pressure on window = 10 bar).

A quite large acceptable domain of cooling parameters appears on Fig. 7. Acceptable parameters can be chosen within the range T_{PbBi} = [250-400] °C and h = [1.5-4.0].10^4 $W/(m^2K)$ for values indicated by Figure 7. Attention should be paid to the fact that strong uncertainties remains, especially concerning the correspondence between h and v along the window and the creep limit.

The frequent beam trips of the foreseen high power protons accelerators will induce fatigue in the structures and especially in the beam window. The risk of crack propagation has been estimated with the help of the RCC-MR design code [2]. Only thermal loading will induced fatigue in the structure as the hydrostatic pressure remains constant. Fatigue is here completely driven by the current density. The number of cycles before reaching crack initiation in the T91 window has been estimated on the basis of the fatigue curves of the T91 steel [3] and the real strain range $\Delta\varepsilon_{eq}$ computed for a conservative thermal cycle: starting from steady-state at nominal power, the beam is switched-off until reaching a new steady-state. The corresponding $\Delta\varepsilon_{eq}$ values goes from 0.010 at 10 $\mu A/cm^2$ to 0.107 at 100 $\mu A/cm^2$.

The fatigue curve of the T91 martensitic steel (Fig. 8) indicates that, even at 100 $\mu A/cm^2$, the number of cycles before crack initiation will be higher than 10^9, which is considerably higher than the average number of beam interruptions encountered, for instance, on the LAMPF for 200 days: about 1.2 10^4 beam cut-off of a duration between 15-20 s. The conclusion is then that frequent beam trips of foreseen high power protons accelerators will not be a limiting factor for the beam window lifetime.

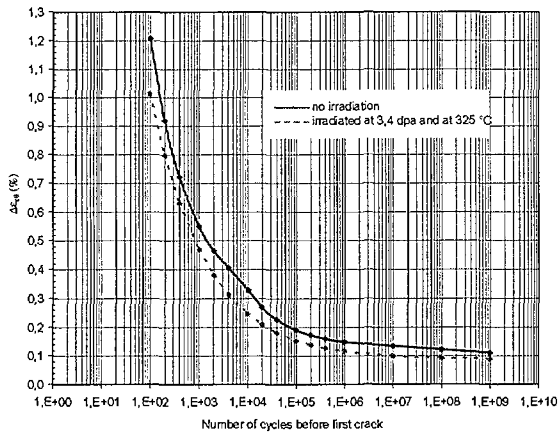

Figure 8: Fatigue curves of T91.

The irradiation effects at such dose on the thermal and mechanical properties have been presented in [4] for an EM10 martensitic steel and concluded that the calculated temperature and stresses remain almost unchanged (very low swelling). This conclusion is preliminarily applicable for T91 as both materials - both 9Cr martensitic steels - stay in the same batch with regard to irradiation.

3.3 The CEA solid spallation target

If the evaluation of the liquid metal reference spallation module shows that it could answer the points required in a ADS dedicated to waste transmutation (neutronic, thermal and mechanical performances), it has appeared interesting to examine the alternative concept, in which the target is made of solid materials. Among all metals we can choose (tantalum, platinum, tungsten, osmium...), tungsten has been preferred thanks to its high temperature of fusion (T_{fus}=3410°C), its density of 19.3 and its thermal conductivity (120 $W.m^{-1}.K^{-1}$ at 500°C). The coolant considered is helium, because of its good thermal capacity (5182 $J.kg^{-1}.K^{-1}$), its good properties regarding to nuclear reactions and to corrosion problems. The pressure of helium has been fixed to 60 bar.

Several concepts of tungsten targets have been studied [5]. We have shown that for a 8.33 mA beam (proton current density of 47 $\mu A/cm^2$) and when the target is made of bored tungsten disks cooled by helium, the 1.2 GeV protons impact leads to a temperature of more than 1000°C. When the target is made of full tungsten disks, we obtain a volumic heat deposition at the center of the first plate of 2300 $W.cm^{-3}$ and a temperature of 2240°C, i.e. $0.66T_{fus}$, which illustrates the impossibility to use massive plates for solid spallation targets. As a result, the best technological option is made of solid particles (cf. Fig. 9). In this case, the ratio surface/volume enables a better heat transfer but the void fraction is harder to adjust, particles suffer from abrasion and erosion entailed by their movement and the cooling speed is limited. In the modelised target, the diameter has been set to 18 cm and the protons beam radius to 15 cm in order to have a current density of 47 $\mu A/cm^2$.

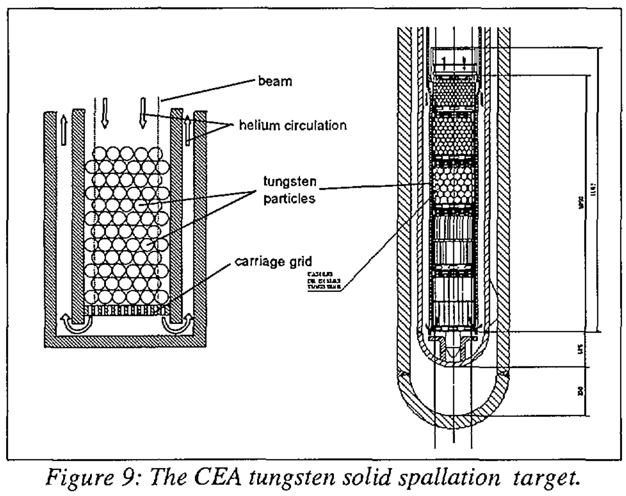

Figure 9: The CEA tungsten solid spallation target.

After calculation, it has been shown [5] that the pressure drop in the case of single diameter particles was almost three times higher than in a case of increasing diameter particles, which would increase as well the power of the helium pumps. As a result, we have chosen to build the target with particles whose diameter go from 1.5 to 8 cm. In this case, the temperature in the center is between 550°C and 800°C (cf. Fig.10), and the temperature in surface of particles is between 500°C and 600°C, which assures that tungsten remains in its ductile zone. This optimisation reduces also the velocity of helium, which lessens technological problems.

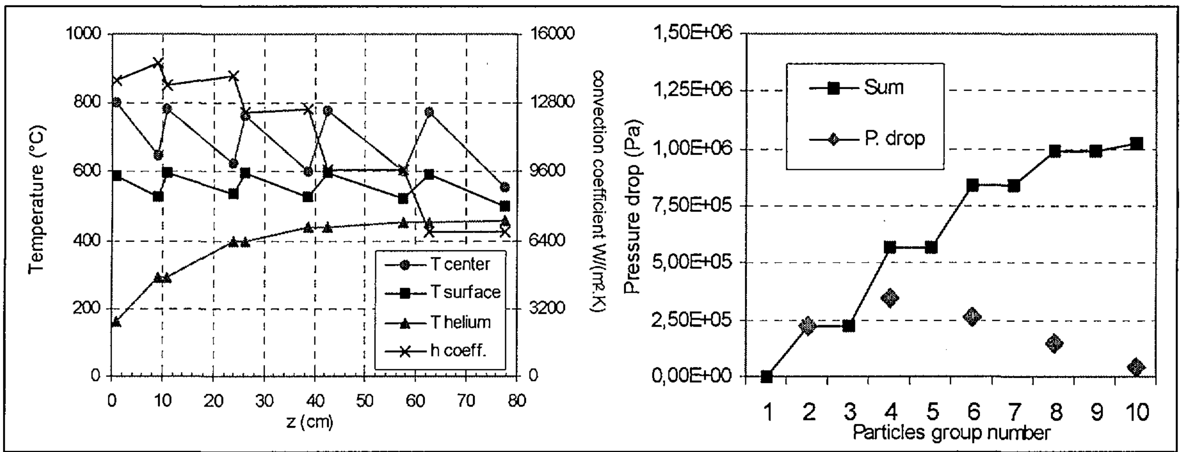

Figure 10: Temperature and pressure drop for a target with particles of increasing diameter.

The *SPARTE* calculations [5] of this target have been made with 1.2 GeV protons and a current density of 47 μA/cm^2. The structural material is T91 martensitic steel (9% Cr). This study gives ratio neutrons/incident proton of 25 with a 8.3 mA, which is 20 % smaller than with liquid target. The efficiency is better because 93.6% of the produced neutrons leave the module. We have noted that 95% of these leaving neutrons have an energy between 5 keV and 20 MeV and that the spectrum corresponding to a tungsten particles target is stronger than the spectrum of a massive tungsten target. The former target enables also to flatten the neutrons flux leaving the module (cf. Fig. 11). So, as in the case of the liquid metal target, this target looks interesting in an incineration purpose design because the strong spectrum will improve the efficiency of the fission reactions and the axial flatness of the flux will assure a good burn-up distribution.

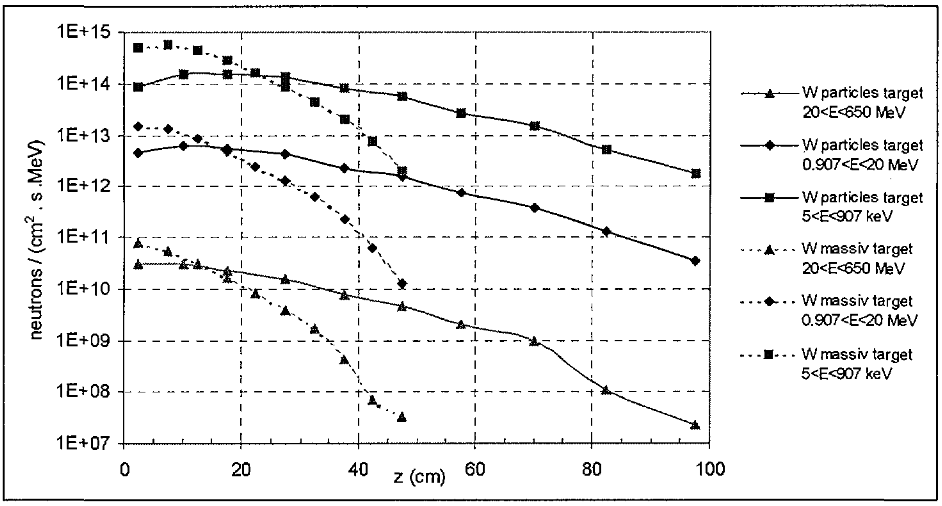

Figure 11: neutrons current density in both concepts of spallation targets.

As we have done for liquid target, when we examine the consequences of irradiation on structural materials, we see that the dpa/EFPD (Equivalent Full Power Day) is the same in the impacted grid of this target and in the window of the liquid metal target, as well as the heat deposition or the gas production (helium and hydrogen). Therefore, these points will not raise major problems concerning the behavior and the lifetime of irradiated structures.

4- DISCUSSION

In the goal of studying ADS dedicated to nuclear waste transmutation, this work has focused on the examination and the design of the *spallation module*. From a look on the main possible options it appears that we have to pay attention to two possible concepts of modules: with liquid or solid metal target. So, CEA has studied a liquid PbBi target and a solid tungsten target.

In the first option, the dimensions are now known and the capacities of crucial components like the impacted window (in T91 steel) to resist to thermomechanical and irradiation effects have been proved, along its whole lifetime (including fatigue phenomena). Such targets are really efficient to produce a neutrons current, and its associated fast spectrum, well adapted to the incineration purpose of the sub-critical core. The thermohydraulic study has enabled to choose a circulation flow inside the module and to optimize the window profile. These evaluations lead CEA to identify an acceptable domain among all the design criteria.

In the solid target option, the feasibility of the concept is proven even with protons density as high as 50 $\mu A/cm^2$ if the target is made of particles. The tungsten particles will necessarily have to be separated in groups of increasing diameter to improve the axial distribution of the neutrons current and the n/p ratio, which in this case, are better than those obtained with the liquid target.

In the framework of this study of spallation targets, several points remain unknown. The effects of an neutrons+protons irradiation on T91 steel, the thermohydraulic correlation along the window, the PbBi flow modelization, or corrosion problems will have to be examined. Further studies will also have to consider accidental and incidental situations (loss of helium pressure...), the necessity to clad the tungsten particles to strengthen their mechanical resistance under irradiation or some lifetime evaluations.

As a result, at this stage of studies both concepts of spallation target present advantages and drawbacks. The liquid target, which remains the CEA reference concept, even if several technological problems are still standing, offers more possibilities regarding to high protons current densities, which is interesting for an extrapolation to an industrial facility. Nevertheless, thanks to all knowledge gathered with HTR studies, the solid target would be developed with less difficulty and its axial neutrons current distribution is more easily adjustable, which appears really interesting if our purpose is to incinerate nuclear wastes. Further studies should reduce the range of the acceptable operating systems parameters and will enable to make the best choice for an ADS dedicated to transmutation.

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