

## Nano-micro-hetro-structured spallation thin target

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### Abstract

*The efficiency accelerator-driven systems are applied to subcritical nuclear structure for energy production, burn out dangerous isotope fission by-products and minor actinides are given by the spallation neutron source.*

*The actual thick spallation targets have some advantages from the point of view of the accelerated beam usage, but their neutron extraction is energy intensive.*

*A new nano-micro-hetero-structured nuclear fuel spallation source is in early development stage. Simulation and theoretical evaluations show that it may bring significant advantages over the actual neutron source and subcritical structure making the energy balance strongly positive and making the accelerator-driven systems an efficient process able to deliver both neutrons and electricity. The neutron target is optimised to harvest the energy of the high yield accelerated particles only, recovering the energy deposited by the beam that did not open any spallation reaction, as electricity. High-energy neutrons are driven through a customised structure that maximises the neutron amplification by its optimal fission cross-section. The driven nuclear structure, usually a subcritical reactor, amplifies the energy of the generated n and converts it into energy and transmutation products in the specialised blanket channels. Enhanced spallation neutron generation uses an accelerator-storage ring system that may be used to pulse a supra-critical reactor structure generating high neutron fluxes, and high electric pulsed power needed in various applications.*

## Introduction

Future nuclear power developments are based on accelerator-driven subcritical system (ADS) foreseen as being of great benefit to the environment and as a potential source of nuclear energy. The rapid increase in the world's economies requires more energy, and nuclear option seems to be the most reliable.

A spallation neutron source is an important element for the majority of ADS applications. The main goals of ADS are transmutation and electric energy production. In a nuclear assembly the produced energy is direct proportional to the number of fissions that depend on the number of neutrons and the material configuration with emphasis on nuclear reaction cross-sections. Presently the criticality condition is applied to fissionable structures in order to make continuous nuclear fission process possible at a certain power level without injecting any external neutrons. It is the cheapest way of obtaining enough neutrons to carry on the fission reaction, but has the drawback of requiring a complicated control mode through the intermediation of the so-called effective criticality. This represents the combined effect of many factors that must be anticipated all the time and considered in every modification meant to maintain the reaction under control.

To produce the fission energy without having the critical mass, it is necessary to supply the necessary amount of neutrons to generate the desired power by fission.

The actual capability of the neutron sources is usually under  $10^{14}$  n/cm<sup>2</sup>-s, while the energetic cost per neutron generated by other means than fission in critical assemblies scales over 50 MeV/neutron.

The power performance obtained in an actual nuclear reactor proved impossible to reach if the criticality is made smaller or eliminated from the process. The trade-off is represented by hybrid structures containing an autonomous source of neutrons and a near critical structure adjusted to a subcritical value larger than 0.94. This configuration brings no cost benefit, being more complicated than the initial critical nuclear reactor since it adds an accelerator system to its configuration.

From almost all the available processes for producing neutrons it turns out that only the accelerator-driven spallation exhibits the necessary performance to be used as a high intensity neutron generator. This kind of ADR is required by the need to produce fuel breeding and fission products transmutation in fast spectrum reactors. In configurations where the delayed neutrons are missing, the system also exhibits variable, hard to predict criticality, making control very difficult for normal devices presently used in thermal nuclear reactors.

## Spallation targets

ADS performance depends strongly on the characteristics of the spallation neutron source (SNS). From the engineering point of view, the basic problems of this kind of application are spallation neutron yield, energy deposition, radiation damage and radioactivity accumulation. Usually, the design of a spallation neutron source starts with an evaluation of the preliminary performance done by computer simulation codes, benchmarked where possible by experimental values.

The well-known high-energy transport code (HETC) also known as LAHET, or GEANT, FLUKA is used in combination with a neutron transport code such as MCNP, MORSE, SNSP, SHIELD, CED/DEM. Some of the groups are using several codes in parallel and are comparing the results [1].

Most of the solid spallation targets are made of Pb or W.

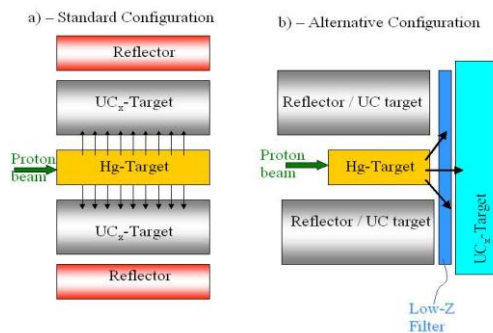
The design calculations performed by the European Isotope Separation On-line Radioactive Ion Beam Facility Design Study (EURISOL-DS) group [2] were done using Hg and lead-bismuth eutectic (LBE) liquid metal targets.

Sensitivity studies in different scenarios were simulated using the Monte Carlo code FLUKA to assess the impact of the projectile particle energy on the neutronics and energy deposition in the spallation target [2]. Optimal target dimensions were also studied for every case as well as the proper target material for the liquid-metal proton-to-neutron converter, since mercury and lead-bismuth eutectic are both reasonable options. Other aspects such as the effect of the beam width on the power densities were studied and a comparison between protons and deuterons as primary particles was performed.

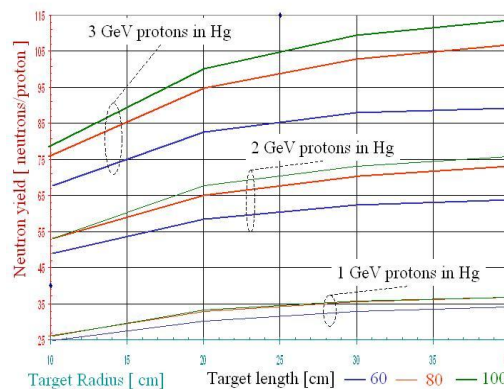
The results showed the benefit of using protons as primary particles and increasing their energy up to 2 GeV, with a beam shape  $\sigma \sim 15$  mm Gaussian distribution on a 15 cm radius and 50 cm long target, in order to reduce the high-power densities occurring in the first few centimetres downstream from the interaction point. The proton beam offers a suitable trade between increasing the neutron and fission yields and reducing the power densities in both the liquid metal and fission targets.

The main configurations for the actual n-spallation sources are given in Figure 1. The standard configuration [Figure 1(a)] contains a cubic-cylinder Hg container with radius  $R$  and length  $L$  in cm, having a strong cooling system made by Hg recirculation. The spallation neutrons dispersed all around the target are amplified in a  $UC_x$  secondary target which produces fission with the energetic neutrons and are confined by a reflector in the forward direction producing a high neutron flux inside. It has been observed that the end-of-range protons do not make a significant contribution; therefore, the Hg spallation target was shortened and the amplification configuration enhanced as is shown in Figure 1(b). Therefore, a part of the beam power is removed by a secondary cooling system, overlapping with an  $UC_x$  target. Figure 2 shows the neutron yield obtained for various radius and lengths for the Hg target, using various energy proton beams.

**Figure 1: Spallation target configuration**



**Figure 2: Geometry, energy neutron yield**



It was shown [2] that 1 GeV protons are completely stopped in less than an 80 cm Hg target, being no significant increase for longer target, while the effective straggling remains smaller than 25 cm. It is also shown that a 3 GeV proton beam requires a bigger target than  $\varnothing 80 \text{ cm} \times 100 \text{ cm}$  length, delivering up to 115 n from a total possible of 120. Deuterons on solid  $^{238}\text{U}$  thick target have shown a slight advantage due to a better cross-section over 55 MeV, and almost double that for protons at 130 MeV.

The overall neutron flux distribution, where the shape of the distribution is about the same with higher values of neutron flux, appear in the periphery of the LBE target due to the lower LBE capture cross-section compared to Hg.

The most significant discrepancy is found in neutron balance densities. A net neutron absorbing region in Hg is obtained for a radius greater than 12 cm and a length greater than 70 cm, whereas the whole target volume has a positive neutron balance when LBE is used due to the fact that no parasitic captures occur in the LBE target. This improvement in the neutron efficiency is due to the significantly lower n-capture cross-sections of the isotopes forming LBE (i.e.  $^{209}\text{Bi}$  and  $^{204}\text{Pb}$ ,  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$ ,  $^{208}\text{Pb}$ ) compared to those forming natural Hg (i.e.  $^{196}\text{Hg}$ ,  $^{198}\text{Hg}$ ,  $^{199}\text{Hg}$ ,  $^{200}\text{Hg}$ ,  $^{201}\text{Hg}$ ,  $^{202}\text{Hg}$ ,  $^{204}\text{Hg}$ ). This neutron self-absorption is an important factor for the epithermal neutron yield and is smaller for LBE.

Another critical issue for high-power spallation sources is the energy deposition in the different elements of the target. The EURISOL Multi-MW target must sustain a 4-5 MW beam [3].

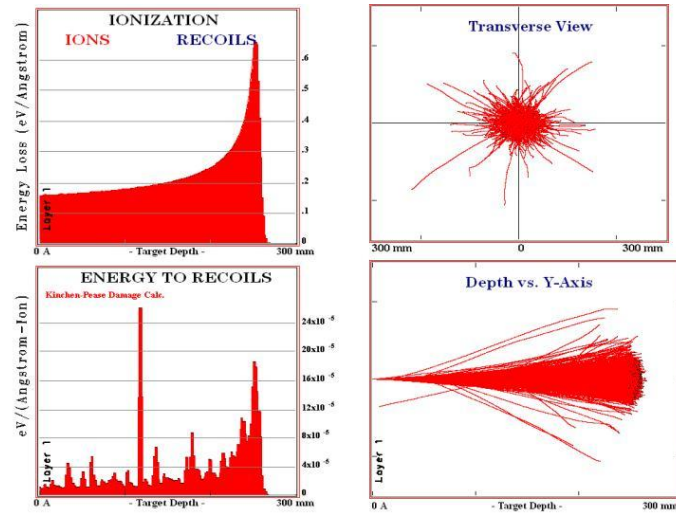
The limitation occurs in the heat flow transport through the lateral surface of the target, finally obtaining a linear increase with the beam and target dimension. The operating conditions are reached when the power density falls inside the acceptable safe limits for the target materials [4].

There are some novel ideas for using a fission target; natural uranium [5] surrounded by a neutron reflector seems useful to obtain larger fission yields [6]. The drawback of lead-bismuth eutectic as a neutron converter material comes from its greater technical complexity (heating assistance required over its melting point at 125°C and the production of important radiotoxic isotopes through irradiation), unless the maximum power density cannot be maintained well below the boiling point of mercury of about 357°C. The use of deuterons makes the neutron yield increase by 15% and the maximum power density by 30%, increasing the costs of a deuteron machine. Beryllium oxide is a good choice to partially recover the escaping neutrons.

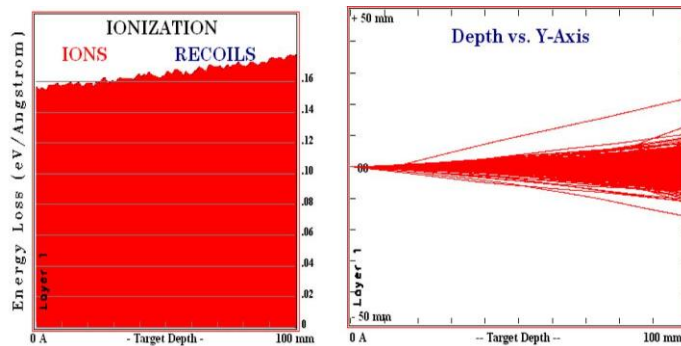
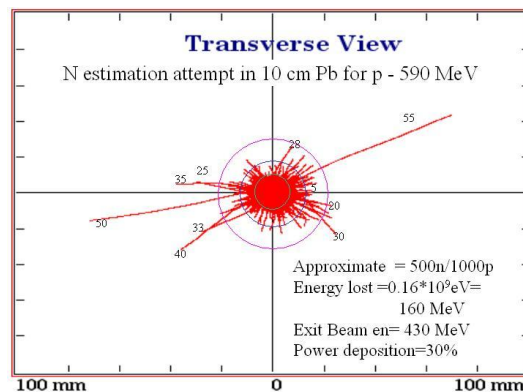
### Thin spallation targets

Following the detailed analysis made for various spallation sources one may realise that in the last part of the range the neutron yield is small while the energy deposition remains significant. In the upper-left, Figure 3 shows the ionisation energy deposition. It shows in this SRIM [7] simulation that the proton range is 25.4 cm, with a straggling of 0.95 cm. The lower-left of Figure 3 shows that the collision with target's nuclei is random, and the energy is significant. The higher the energy transfer to recoil is, the higher the probability to generate a high neutron yield spallation. The lower-left of Figure 5 shows about six high yield spallation reactions for 1 000 particles in the first 10 cm of the target.

In the right side of Figure 3 is shown the trajectories of the stopping protons in the target in transverse and lateral view. The only highly deflected ions are those producing neutrons. If six strong interactions produced in the first 10 cm we may say that about 90-100 neutrons have been generated by 1 000 protons from an estimated total of about 250 neutrons.

**Figure 3: Protons of 590 MeV stopping in 30 cm solid Pb**

In the left of Figure 4 is shown that the energy lost in the first 10 cm of Pb by a 590 MeV proton is about 160 MW (representing 30% of the beam energy) to produce about 45% of the neutrons. The image in Figure 5 shows a way of estimating the spallation. It can be seen that the target may be further reduced to get the optimum between the energy lost in target versus the number of neutrons generated. This optimisation shows that a thickness between 2-6 cm may host the optimal value in this case. In reality the optimal value is obtained with more difficulty, using a global optimisation.

**Figure 4: Ionisation energy and trajectories in 10 cm Pb****Figure 5: Proton trajectories transverse view**

### Storage-ring-based spallation systems

The usage of less than 30% of the beam power to produce neutron spallation allows the rest of beam energy and accelerated particles available for being dumped in a larger dump target minimising the cooling requirements, or may be recovered and reaccelerated to the necessary energy and reapplied on target. In this more than two times more neutrons may be generated way with the same energy loss.

Ion beam recovery represents a technological challenge compared to dumping all the beam power in a target and attempting to extract as many neutrons as possible. The electron strip effect makes these action even more difficult if heavier ions, but protons are used. Thin n-target storage ring collider mainly uses the high-energy interactions only providing a high yield per collision, having the n-spectrum displaced towards fast neutrons in the energy range over 15 MeV/neutron.

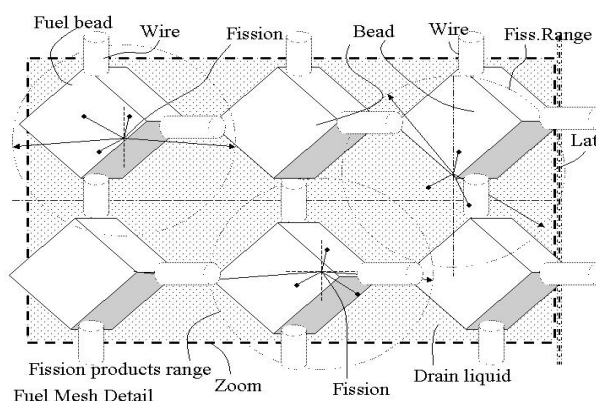
The applications use these micro-nano hetero fuels to increase the neutron flux in a spallation neutron source called a “neutron booster”. The spallation target (similar to MEGAPIE) may be surrounded with fissile layers, making a subcritical system with  $k_{\text{eff}}$  about 0.95-0.99, and increasing the neutron fluxes up to a factor of 5-10. Estimations show that at least 2 kg of fissile material is needed [6]. After passing through the material structure, the beam emittance is increased, and an accelerator focusing structure is recovering the scattered beam, refocusing it and sending back in the storage ring for a new collision with the target. Before collision, a buncher device is applied to prepare the beam shape and timing in order to make the beam recovery easier and more effective. A spallation neutron converter has to remove the heat, produce as many neutrons as possible, with energies less than 10 MeV/neutron, absorb the least possible from the beam energy, and thin enough not to increase the emittance of the beam after passing through the target. One solution is to use a hetero-structure in order to achieve the necessary thermal conductivity and the advantages of a liquid with the best actinide spallation factor.

### Novel spallation targets based on micro-hetero structures

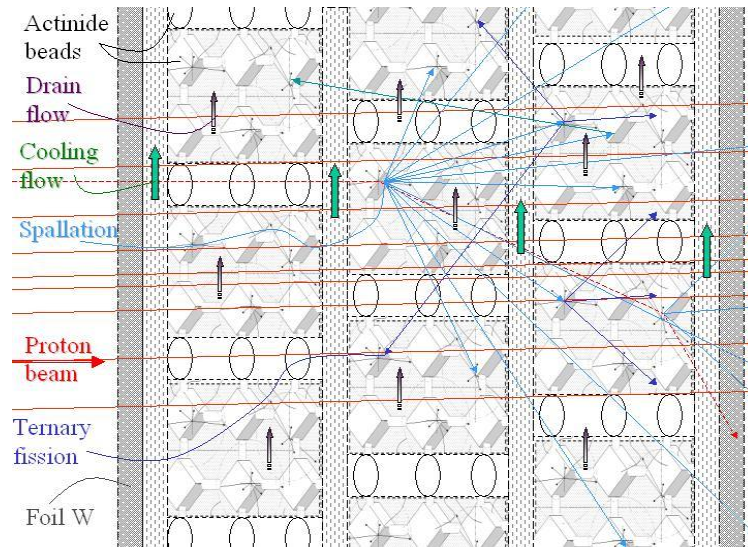
This novel fuel is made from beads of actinide fuel (such as  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{242}\text{Am}$ ) with dimension shorter than the range of fission product, immersed in a drain liquid such as LBE.

The fuel structure presented in Figure 6 is made from beads of actinide compounds with dimensions smaller than the fission products range sunk into a liquid metal as LBE. This makes the fission products deposit their energy into the drain liquid. Figure 7 shows a thin target with dual flow, one cooling LBE micro-flow in parallel sections removing the heat and the other is the drain flow, which has very low speed and washes the actinide beads and carrying away the fission and spallation product leaving the target clean from radioactive fission products and with constant reactivity. Figure 7 also shows a colliding proton that induces spallation similar to the deflected particles shown in Figures 3-5.

**Figure 6: Micro-hetero-structured fuel mesh**



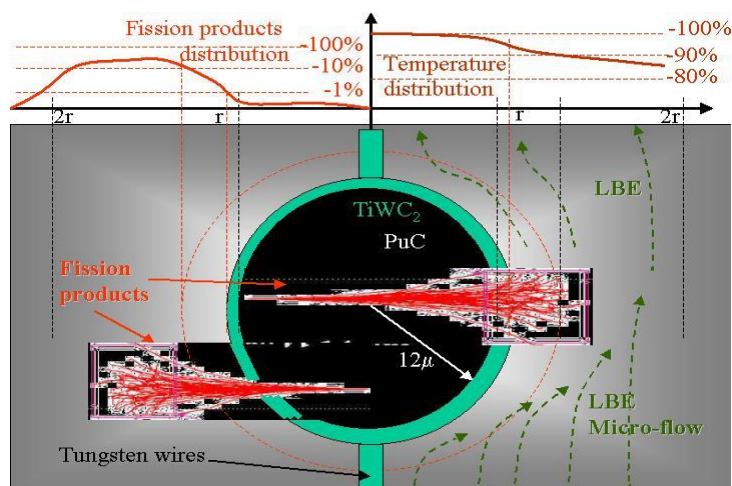


**Figure 7: Hetero fuel thin spallation target**

The generated neutron shower scatters and produces secondary fissions that in turn generates up to 10 neutrons that may produce ternary fissions and so on. Due to the use of a combination of actinides and spallation targets the neutron yield is very high. Depending on beam bunching the n-pulses may be high and intense, but may also be enlarged by the n-amplification mechanism present in near subcritical structures.

The advantage of this type of fuel over the classic liquid or solid spallation proton-to-neutron conversion target is that it has the thermal conductivity of the liquid target and the n-yield of the solid actinide target [8]. Moreover it takes almost no radiation damage and removes the fission products from the active area leaving the reactivity constant. From the energetic point of view, being thin it takes less than 10% of the beam energy yet delivers about 60-80% of the neutrons for the case with total beam dump in a thick target, producing factor of 5 energy improvement.

Figure 8 shows in detail that the thermal spike of the fission reaction is produced outside the bead, depositing the released energy directly in the liquid with the fission product adding a mild temperature increase on the bead's surface that increases its mechanical stability. This is a modified structure of that used in beam targets.

**Figure 8: Detail on fission process inside the micro-bead**

### Nano-structured electronic cooling spallation targets

The micro-hetero-structured target offers a thermal conductivity 50% lower than the pure liquid used, depending on the bead-drain liquid ratio, but several times higher than the thermal conductivity of the ceramic material containing the actinide material. It exhibits power density limitations given by the maximum allowable safe temperatures, which sets the maximum power density at several kW/cm<sup>3</sup>. In order to be efficient, the beam will put a significant amount of power in the structure and the use of a collider storage ring will maximise the current density.

Using electric current as an energy transport agent is a much more effective form of removing the beam energy deposition and is much faster than by thermal conduction and mechanical heat flow transport. The nano-structures that have the capability of direct conversion of the particle's kinetic energy into electricity may do that. Figure 9 shows how a moving particle coming from a fission reaction or from an externally produced radiation converts its kinetic energy into electricity by crossing different material nano-layers. A "Clci" structure is formed by a high electronic density conductive material "C" that may contain actinides too (generically represented in the figure by "High-Z") is separated by an insulator "I" from a low-electronic-density conductive material "i" (generically called "low-z") insulated at its turn and creating a direct conversion cell. When radiation crosses this structure it mainly interacts by ionisation, creating a bigger electronic avalanche in the "High-Z" material that stops in the "low-z" material which loses up to 8 times a smaller electronic avalanche than the "High-Z" material as shown in Figure 10. That means that in the "low-z" materials remain accumulated up to 7 "low-z" avalanches that polarise this material negatively. Connecting the "low-z" materials via an external resistor load to the "High-Z" material the polarisation voltage generates a current that takes out the accumulated energy. The conversion energy may be as high as 7/8, or about 90%. The real conversion efficiency is construction- and beam-dependent and may be smaller than the maximum predicted theoretical efficiency. Finally, it is possible that an important part of the beam deposited energy and the energy released by fission during the neutron amplification can be recovered as electric power, making the thermal stress in the material lower.

In fact, the energy removed as electric current will not take part in material heating, [9] but will generate heat in the electric circuit by the Joule effect. To obtain the two effects simultaneously it is necessary to replace the actinide beads in Figures 7, 8 with the "Clci" nano-structured beads, connected to each other as shown in Figure 11. The voltage will be accumulated and transmitted outside the target to a DC/AC converter. This kind of target will allow higher energy beams to be applied and depending on overall conversion efficiency the power may be recovered and may generate some extra power.

Figure 9: Direct conversion in nano-structures

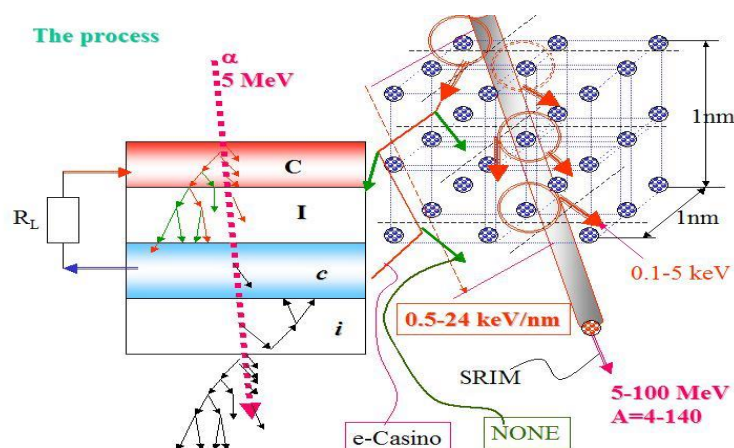




Figure 10: Layer knock-on electron yield variation in direct conversion in nano-structures

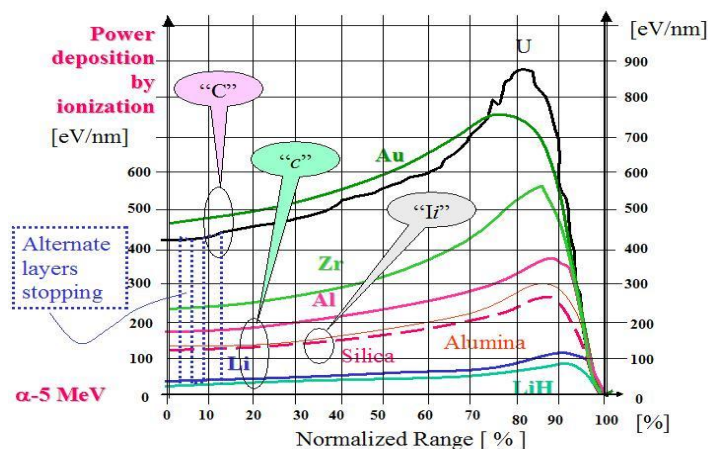
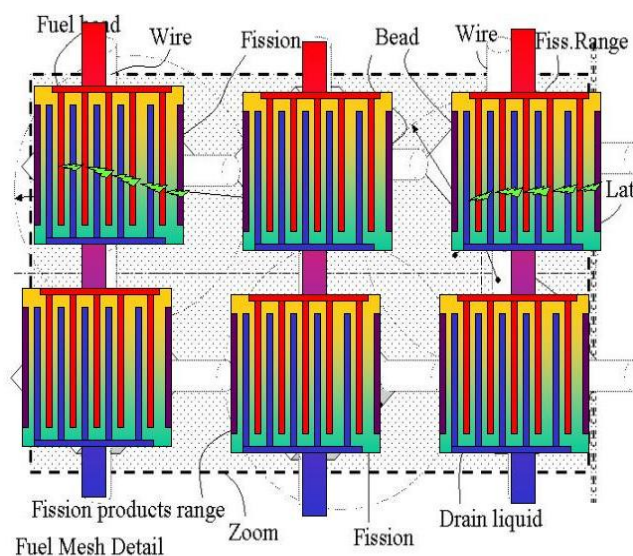


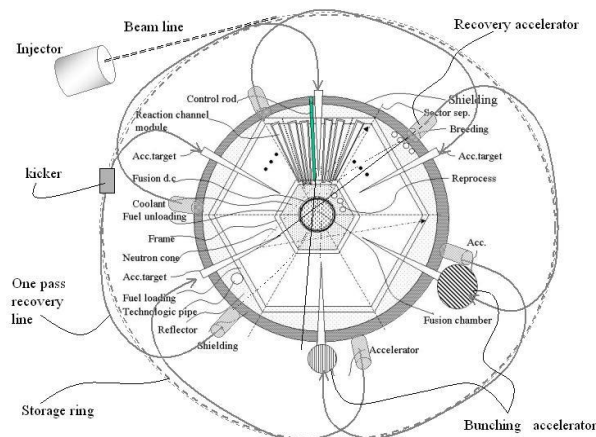
Figure 11: Micro-nano-hetero structure



### Advanced accelerator-driven systems reactors

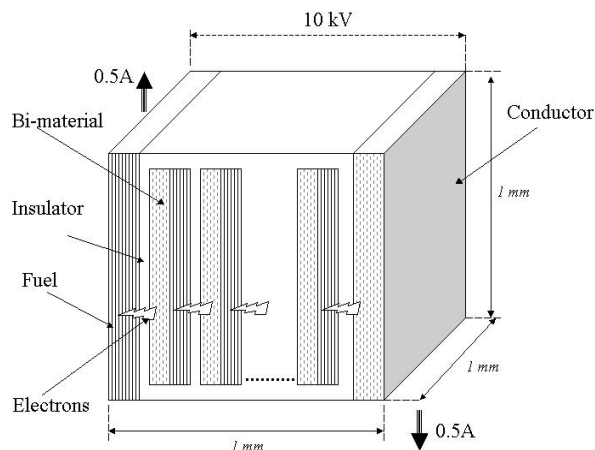
Direct conversion micro-nano-hetero structures may bring some improvement to spallation targets, possibly being able to extract the heat, fission products and electric current as power deposited by the beam and generated by the fission process.

The micro-nano-hetero structures may be applied at each spallation target in the nuclear reactor structure, to the neutron amplification subcritical cavity around the target, and to the nuclear reactor structure as shown in Figure 12. An injector, putting accelerated particles into a storage ring, completes the structure that uses a kicker to deliver the beam into the reactor spallation targets chain [10]. The reactor distribution system is made of an accelerator preparing the particle bunch for the interaction with the spallation target, and an extraction device (similar to an accelerating focusing-bunching structure) recovering the non-reacted beam and restoring its parameters.

**Figure 12: ADR multi-beam structure**

In subcritical structures, depending on the ratio between the beam neutron deposition and the decay of the neutrons in the subcritical structure, the power may remain constant depending on the beam neutron generation or may vary. When the production of neutrons by spallation is bigger than the loss of neutrons as a result of decay in the subcritical structure the power in the nuclear system builds up.

The use of a nano-hetero structure in the future is predicted and will increase the power density from the present  $0.2\text{--}1\text{ kW/cm}^3$  used in nuclear reactors to several hundreds of  $\text{kW/cm}^3$  or more. The theoretical limit is in the domain of a few  $\text{MW/cm}^3$  based on material calculations, but the effective value will be driven by the conversion efficiency of the energy radiation power into electricity. The remaining unconverted energy must not surpass the safety limits of energy removal by heat flow, which is under  $1\text{ kW/cm}^3$  as shown in Figure 13.

**Figure 13: Power density estimation in capacitor structures**

As shown in Figure 13 the trade-off to maximise power extraction is made between sections of the conductive material versus the thickness of the insulating material [11]. The conductive material fraction (and its distribution in connections) limits the acceptable current value, while the fraction of insulator gives the maximum break-down voltage that by internal connections is traded against the capacitance. In the simple calculation case,  $1\text{ mm}^3$  of material is used, capacitive effects have been ignored, and various cross-sections carrying directly generated current have been considered. For a  $0.1\text{ mm}^2$  copper wire cross-section, about  $0.5\text{ A}$  can be transported, and  $0.9\text{ mm}$  insulator may offer up to  $10\text{ kV}$  voltage stand-off, producing a total power of about  $5\text{ kW/mm}^3$ . It is estimated that a superconductive structure could carry at least

10 times more current, making the power a factor of 10 higher. In fact, what matters most is the conversion efficiency. Considering an efficiency  $\eta = 0.9$ , and the heat removal capability of about  $1 \text{ W/mm}^3$ , the total power delivered will be about  $9 \text{ W/mm}^3$ . A little bit less than 1 order of magnitude increase in power from more than 3 orders of magnitude is theoretically possible. With the associated Joule effect this value becomes even smaller. Therefore, to obtain the best parameters these structures have to be optimised accounting for many parameters and operation conditions.

### Pulsed neutron sources

Nano-hetero structures show a potential of operating with increased performance, generating more neutrons at a lower energy cost per neutron. It has the drawback that the accelerator structure gets more complicated, evolving from a beam dump design in hard thermal stress conditions to a thin high yield target in a restoring and recirculating beam system.

It has also been shown that the power enhancement of the nano-hetero structure greatly depends on the direct conversion efficiency obtained for the specific application and the capability of the structure for removing heat. A high-neutron pulse reactor bases its operation on high-power deposition over the capability of heat extraction, but absorbs it in the material's thermal excursion near the upper safety limits. The pulse time may be calculated as:

$$t_{on} = \frac{mc_p \Delta T}{P_{ef}(1 - \eta)} \quad (1)$$

where  $m$  is the mass of the system,  $C_p$  is the specific heat,  $\Delta T$  is the temperature difference between the starting temperature and temperature limit,  $\eta$  is the energy conversion efficiency, and  $P_{ef}$  is the effective power. In actual systems with criticality controlled by mechanical means, the neutron burst must be initiated by radioactive sources, otherwise the time it takes to reach the desired level becomes very long. In ADS, the number of neutrons is dependent on the pulsed power of the beam applied on target. Compared to actual systems, the nano-hetero structures in a spallation-subcritical structure may bring a factor of more than 100 increase in the neutron density by using a shorter time pulse period.

The short  $T_{on}$  is obtained by the high beam pulse application on target and its multiplication in a supra-critical structure, while the shorter  $T_{off}$  is obtained by the neutron deficit left after the beam shutdown. This will allow higher neutron density in the triangular regime or higher quality rectangular neutron pulses.

The actual performance for pneumatic-actuated pulsed reactors such as the Annular Core Research Reactor (ACR) of Sandia National Laboratory gives a peak power of 33 GW equivalent, and a pulse width of 7-7.5 ms for an energy output of 280-310 MJ, with a safety limit at 500 MJ (equivalent of 60 GW). It runs in a steady-state regime in "zero power" mode at 2 MW, and is enhanced to run up to 4 MW [12].

A combination of a pulsed power electromagnetic rail gun, with an accelerator storage ring pulsed beam may produce neutron pulses in the hundreds of microseconds domain and TW equivalent pulsed power regime increasing the neutron flux up to 1 000 times.

In a lower regime with continuous modulation it opens the way towards neutrino communication systems having the pilot modulator performed by the accelerator system.

### Conclusions

The concept of novel micro-nano-hetero structures remains in the early development stage, theoretical evaluation and computer simulations with experimental data coming from collateral experiments. There is no complete experiment that proves the viability of the systems developed as natural applications of the new materials.

The micro-hetero structure is capable of enhancing the neutron yield and may remove the fission products from the spallation target.

The micro-hetero-structure is enhancing the neutron spallation yield, while having a thermal conductivity near to a liquid, and may be built in thin targets.

The direct-conversion nano-hetero structure recovers part of the fission power as electric energy making possible an increase in the power in the target by several times.

The micro-nano-hetero structure makes possible the use of both effects – liquid thermal conductivity and electric power production from direct conversion.

The thin spallation target concept brings some advantages in power reduction and neutron yield, but requires more complicated accelerator storage ring systems.

The application of the accelerator spallation targets to pulsed nuclear reactors may produce pulses well in excess of  $10^{20}$  n/cm<sup>2</sup>/s and pulse-lengths in the microsecond domain.

The development of future reactors using subcritical structure spallation targets and nano-hetero structures may bring significant enhances in safety, transmutation, and energy production.

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