SPECIFIC FEATURES OF PARTICULE/MATTER INTERACTION FOR ACCELERATOR-DRIVEN SUB-CRITICAL REACTORS

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Abstract. Accelerator-driven sub-critical reactors that are envisaged for the transmutation of nuclear waste use intense neutron fluxes produced through spallation reactions in a heavy metal target. The mechanism and specific characteristics of the high-energy nuclear reactions that occurs in ADS are described. The impact on target design, radioactivity production and material damage is discussed. Also, the general features of the simulation tools used to design ADS are presented and the precision with which each quantity important for applications can be predicted is given.

Keywords: Spallation reactions; accelerator-driven systems.

1. Introduction

An accelerator-driven sub-critical reactor (ADS) is a system, in which an intense neutron flux produced through spallation reactions in a heavy metal target is used to drive aa sub-critical core. The spallation reactions are induced by a high-intensity proton beam of energy around 1 GeV. Minor actinides (MA) introduced in the reactor fuel can then be transmuted through capture and fission reactions. The goal being to finally fission the minor actinides, fast spectrum core, in which the ratio of fission to capture is more favorable, are generally preferred as transmuters. The transmutation of fission products, once considered, has been found to be not enough efficient and too expensive and is no longer foreseen. The main advantage of a sub-critical system compared to a critical one is the fact that fuels with a higher content of minor actinides can be used. Indeed, the introduction of MA into a reactor leads to the degrading of the safety parameters as the Doppler coefficient or the delayed neutron fraction. This limits the content of plutonium and minor actinides in a critical reactor but is less a problem if the system is sufficiently sub-critical.

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Materials Issues for Generation IV Systems

One of the objectives of GEN-IV nuclear systems is to minimize and manage their nuclear waste. A way to reduce nuclear waste is to transmute the minor actinides, after separation from the plutonium, which can be reused, and fission products that will go the storage. Two options are envisaged: either homogeneous re-cycling, in which a small amount of MA is transmuted in all the reactors of the park without degrading too much their efficiency and safety, or a double strata scheme, in which dedicated transmuters burn the waste arising from several commercial reactors. In the latter case, ADS are certainly promising since they could accommodate high contents of MA, as above explained, and therefore only a limited number of ADS would be needed. A possible deployment scenario has been for instance proposed by EDF, in which, in a park of fast reactors producing 60 GWe, ADS corresponding to 1.76 GWe would be needed to transmute the MA (Grouiller 2006 et al.).

2. Spallation reactions

2.1. DEFINITION

The specificity of particle-matter interactions in ADS comes from the occurrence of spallation reactions in the neutron-producing target. A spallation reaction is defined as the interaction of a high-energy (above ~100 MeV) nucleon with a nucleus leading to the emission of light particles, mostly neutrons, and leaving a heavy residue. This process has actually been known for a long time, since it was first observed as particle cascades in cosmic ray interactions. Already with the first accelerators, reactions were found in which the target nucleus looses a lot of nucleons. Serber (Serber 1947) then proposed to describe spallation as a two step mechanism, description which is still used nowadays: the first stage can be viewed as a succession of individual nucleon-nucleon collisions which leads to the ejection of a few energetic particles, including some pions created in inelastic collisions, and to the deposition of part of the incident energy into the target nucleus. The excited remnant then decays by evaporating particles, mainly neutrons or by fission in the case of heavy nuclei. Some of the nucleons ejected during the spallation reactions have sufficient energy to induce further spallation reactions with neighbouring nuclei inside a thick target, leading to a multiplication of the emitted neutrons. Typically a 1 GeV proton on a lead target yields around 20 neutrons. It is therefore possible to produce with high-energy proton beams intense neutron fluxes that could serve for various applications.

2.2. OTHER APPLICATIONS

Spallation neutron sources can be used for condensed matter and material structure studies. The fast neutrons produced by spallation are then moderated in a light or heavy water tank and reflectors are used to direct neutrons into beam tubes. The only continuous source is SINQ at PSI, Zurich, where the first Pb-Bi eutectic liquid target (Bauer 2001 et al.) has recently been successfully tested. New pulsed sources are under construction as SNS in USA, or JSNS in Japan. The European ESS project is still under discussion.

Fundamental research in nuclear physics and astrophysics is demanding a new generation of high intensity radioactive ion beams (EURISOL project), with an emphasis on neutron-rich isotopes. High intensity proton or deuteron accelerators are foreseen to produce neutrons, directly or through a light element converter, that could induce fission reactions in an uranium block and then, after extraction and separation, provide neutron-rich fission fragment beams.

Cosmic rays are composed mostly of protons and alpha particles with an energy spectrum presenting a maximum near 1 GeV/A. Cosmic rays bombardment of spacecrafts is responsible for severe problems of radiation damages, especially in computer electronics. Spallation reactions are also important to explain part of the elemental abundance distribution in galactic cosmic rays.

2.3. SPALLATION REACTION MODELLING

Simulation codes used for spallation source design generally consist of the coupling of a high-energy transport code, which handles the transport and interactions of the incoming proton and all the produced particles down to 20 (sometimes 150) MeV, and a low-energy neutron transport code utilizing evaluated nuclear-data files below. In the high-energy transport codes, the elementary cross-sections and characteristics of all produced particles are calculated by nuclear-physics models. To provide reliable predictions for quantities related to spallation reaction it is therefore necessary to have models describing correctly all the features of the reactions, validated on appropriate experimental data. During the last years, new high-quality experimental data have been collected, leading to a better understanding of the spallation reaction mechanism and allowing the testing of the currently used high-energy models (HINDAS 2005). This has resulted in the development of more reliable spallation models, as the INCL4 (Boudard et al. 2002) - ABLA (Junghans 1998 et al.) combination of intranuclear cascade and de-excitation models in Europe or CEM2k in USA (Mashnik et al. 2005), both now available in MCNPX (Hendricks et al. 2003). In this paper, we will try to give, when possible, the degree of reliability that can be expected from the model predictions.

3. Specificities of spallation reactions

3.1. NEUTRON PRODUCTION

In an elementary spallation reaction, high energy nucleons are ejected during the cascade stage. Their number is rather small, around 2 neutrons and 2 protons above 20 MeV per reaction in the case of Pb at 1 GeV, but they carry out the largest part (about 90%) of the energy and induce secondary reactions in a thick target. During the de-excitation of the excited remnant nucleus low energy (below 20 MeV) particles, mostly neutrons (around 15 in Pb) are emitted. The energy spectrum of neutrons produced in an elementary p+Pb reaction at 1 GeV is shown in Figure 1a. In an ADS, the high energy tail of the neutron spectrum can reach the fuel so it is important that simulations are done globally and do not assume that the spallation target can be decoupled from the rest of the ADS. Also, for radioprotection, these high energy neutrons should be properly taken into account. Although very few, the produced π^0 should be considered since they decay into very high energy (70 MeV) photons.

Because the reaction cross-section of a high energy proton is large (1.7 barns on Pb) a large fraction of the primary proton beam interacts in the first centimeters of the spallation target before slowing down. This is illustrated in Figure 1b (full line), which shows the respective numbers of reactions per incident proton as a function of the energy of the reaction for the different types of particles involved in the interaction of a proton with a thick target. The large number of low energy interactions is due to secondary particles and is dominated by neutron reactions because low energy protons have a large probability to stop without interacting. It can be noticed that pions also induce secondary reactions. All this explains that the maximum of the energy deposition in a spallation target is located close to the entrance of the beam, as it can be seen in Figure 2 in the case of the MEGAPIE target.

A spallation target should be optimized to produce the maximum number of neutrons. Heavy elements, which lead to large numbers of evaporated neutrons, are chosen. Actually, there is very little difference between the different possible materials in terms of neutron production, as illustrated in Figure 3 from Letourneau et al. (2000), that shows the average neutron multiplicity per incident proton in different materials as long as the target thickness is sufficient to reach the plateau observed in Figure 3a.

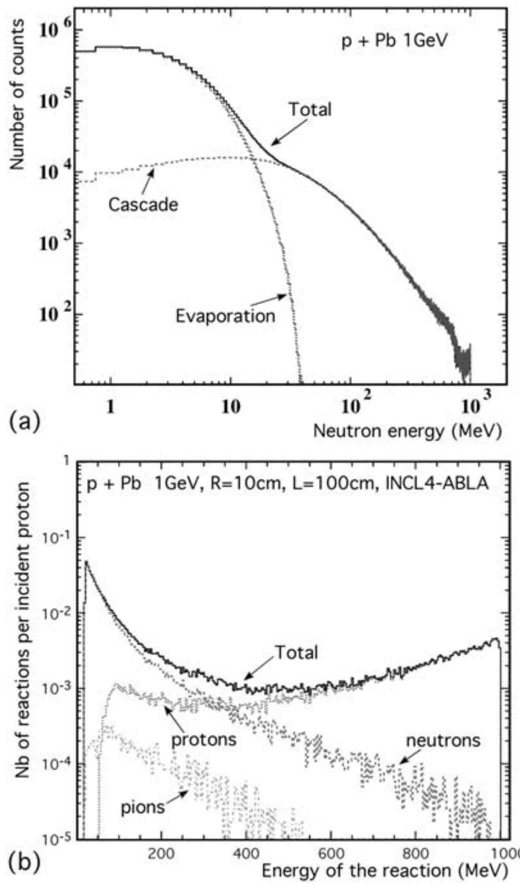

Figure 1. a) Energy spectra of the neutrons produced in the INC and de-excitation stages in the p+Pb reaction, b) Respective and total numbers of reactions per incident proton due to the interactions of the different particles involved in a thick Pb target bombarded by a 1 GeV beam.

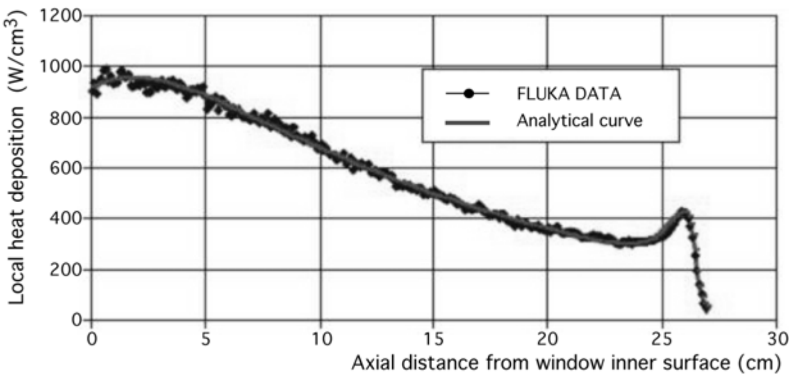

Figure 2. Calculated local energy deposition along the beam axis in the MEGAPIE target. From Kirchner et al. (2003).

In fact, the choice of the target material is mostly dictated by technological reasons. Generally, liquid targets (Hg, Pb, Pb-Bi eutectic) are preferred to solid ones because the necessity of a cooling system reduces the effective density of (Bauer 2004 et al.). It can be seen that solid W or Ta cooled by D_2O produces significantly less neutrons than a liquid Hg target. Recently, the replacing of the standard SINQ target, made of Pb rods inside zircalloy cladding, by the liquid Pb-Bi eutectic MEGAPIE target has been found to increase the flux available to the neutron users at PSI by a factor 1.7 (Zanini et al. 2007). In addition, the liquid metal is its own coolant, the target does not suffer from radiation damage and volatile spallation products can be removed during operation. However, liquid targets have important constraints as the necessity of a container with, in particular, an entrance window for the beam, which is subjected to important radiation damage. Also the corrosion by the liquid metal of the structural materials is a matter of concern, as well as the highly activated circulating fluid. Solid targets (Ta, W, depleted U) have the advantage of simplicity (no container, spallation products confined) but have a limited power evacuation and are subjected to radiation damage.

As regards the optimum beam energy, the relevant variable is the number of produced neutron per GeV in the beam. Several measurements (Letourneau et al. 2000) have shown that this number increases with increasing up to 1-2 GeV above which it tends to decrease. This is due to the opening of many meson production channels that carry out part of the proton energy without creating neutrons.

Most of the available spallation models generally very well predicts total number of produced neutrons as well as their energy and angular distributions.

3.2. LIGHT CHARGED PARTICLE PRODUCTION

Reliable estimations of light charged particle production are important for spallation neutron sources: for instance, helium is a concern in structural materials, in particular the window separating the accelerator vacuum, because it can lead to swelling and embrittlement; tritium can escape from the liquid target and cause problems of radioprotection. The production rate of light charged particles increases with increasing incident energy up to a plateau around 1 GeV as can be seen in Figure 4 from Filgeset al. (2001). This means that production rates are much larger than in a conventional reactor. For instance, 2200 appm He per year are expected in the proton beam window of JSNS (Watanabe 1998) to be compared for instance to 250 expected in the first wall of a fusion reactor. DPAs due to the recoiling nuclei are also important (21 DPA/year are found in the same paper).

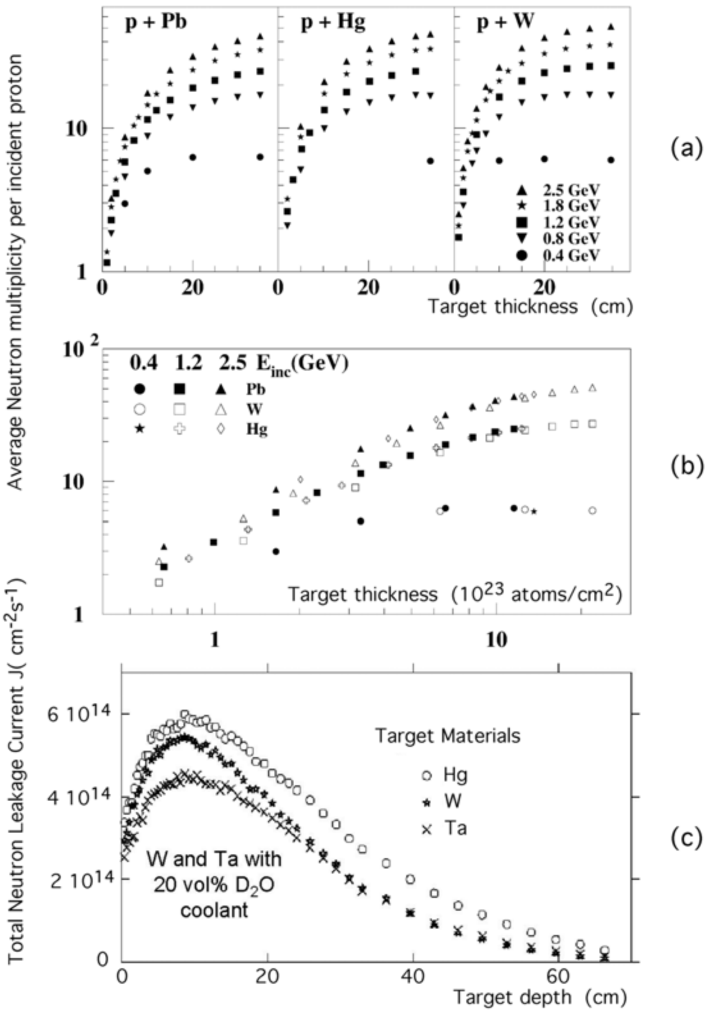

Figure 3. Average neutron multiplicity per incident proton for different materials and different beam energies as a function of the target thickness in cm (a) and in atoms/cm^2 (b). From Letourneau (2000), c), Comparison of the neutron current produced in a liquid Hg target and in solid W or Ta targets cooled with D_2O as a function of the target depth. From Bauer et al. (2004).

Up to now, light composite particle production was rather poorly predicted by the different models implemented into MCNPX (Rapp et al. 2006) or by FLUKA (Pienkowski et al. 2006). Generally, in the models only emission during the evaporation stage is considered. In light composite particle energy spectra two different components are generally observed: a low-energy isotropic one coming from the evaporation stage, and a high energy tail more forward peaked. An attempt to separate the two components has been done recently by Herbach et al. (2006).

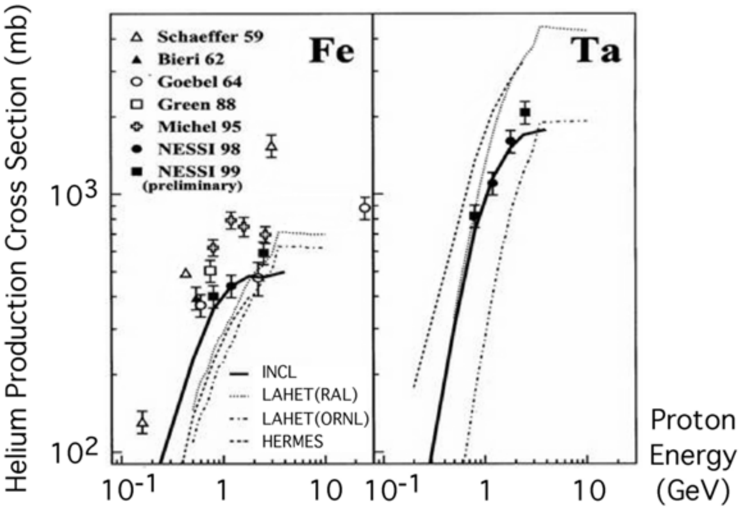

Figure 4. Helium production cross-sections in Fe and Ta as a function of bombarding energy measured and calculated with different models. From Filges et al. (2001).

Figure 5 is taken from Herbach et al. (2006), and shows the relative contribution of this high-energy component, called pre-equilibrium in this paper, relative to the total yields of the different light composite particles, versus the target charge, at 1.2 GeV. It can be seen that, while ^{4}He is produced predominantly by evaporation, for the other light composite particles the so-called pre-equilibrium contribution is far from being negligible, reaching 40% for tritium and even 60% for ^{3}He on high Z targets. The level of this contribution in the production cross-sections of tritium and ^{3}He shows the importance of being able to account for the high-energy tail with the models. This is the reason why the possibility to emit light composite particles through a mechanism of coalescence in phase space has been introduced in a new version of INCL4 (Boudard et al. 2004).

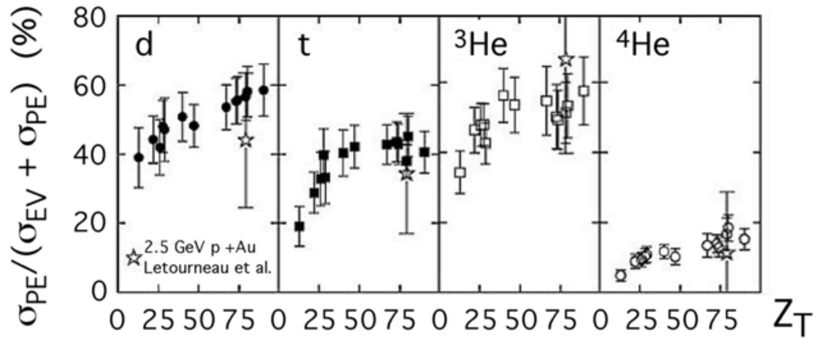

Figure 5. Contributions of the pre-equilibrium emission relative to the total yield of light composite particles. The data measured for reactions with 1.2 GeV protons are plotted as a function of the atomic target number Z_T. From Herbach et al (2006).

The ABLA model in his standard version only allows the evaporation of n, p and ^4He. This means that tritium cannot be reliably predicted with INCL4-ABLA. This is a major deficiency as tritium appears, for instance, to be a major contributor to the radioactivity of the MEGAPIE target around 10 years after irradiation (Lemaire et al. 2007). In order to solve this problem, ABLA has been recently modified to include the evaporation of all types of light particles and emission of intermediate mass fragments (Ricciardi et al. 2007) with appropriate emission barriers.

Figure 6 shows the result (solid line) obtained with the new versions INCL4.4 coupled to ABLA07, for total helium production cross-section in iron, important for material damage studies, as a function of proton incident energy. The comparison to the experimental data and to calculations with other models shows that obviously, the new models represent a progress compared to the previous one (dashed line) or to Bertini-Dresner (dotted line). Tritium production, not shown here, is now also reasonably predicted, the non-evaporative part coming from the INC stage being probably a little overestimated (Leray et al. 2007).

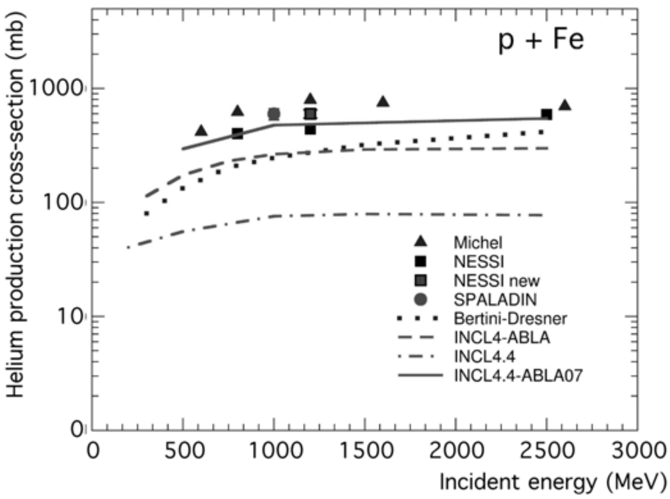

Figure 6. Helium production cross-sections in iron calculated with the new versions INCL4-ABLA07 compared to data measured by different groups and to calculations with the standard INCL4-ABLA and Bertini-Dresner models. The dashed-dotted curve is the contribution from the cascade cluster emission. From Leray et al. (2007).

3.3. RESIDUE PRODUCTION

An important feature of spallation reaction is the large number of different residues that are produced. The example of proton on lead at 1 GeV, measured at GSI using the reverse kinematics technique (Wlazlo et al. 2000, Enqvist et al. 2001), is shown in Figure 7a.

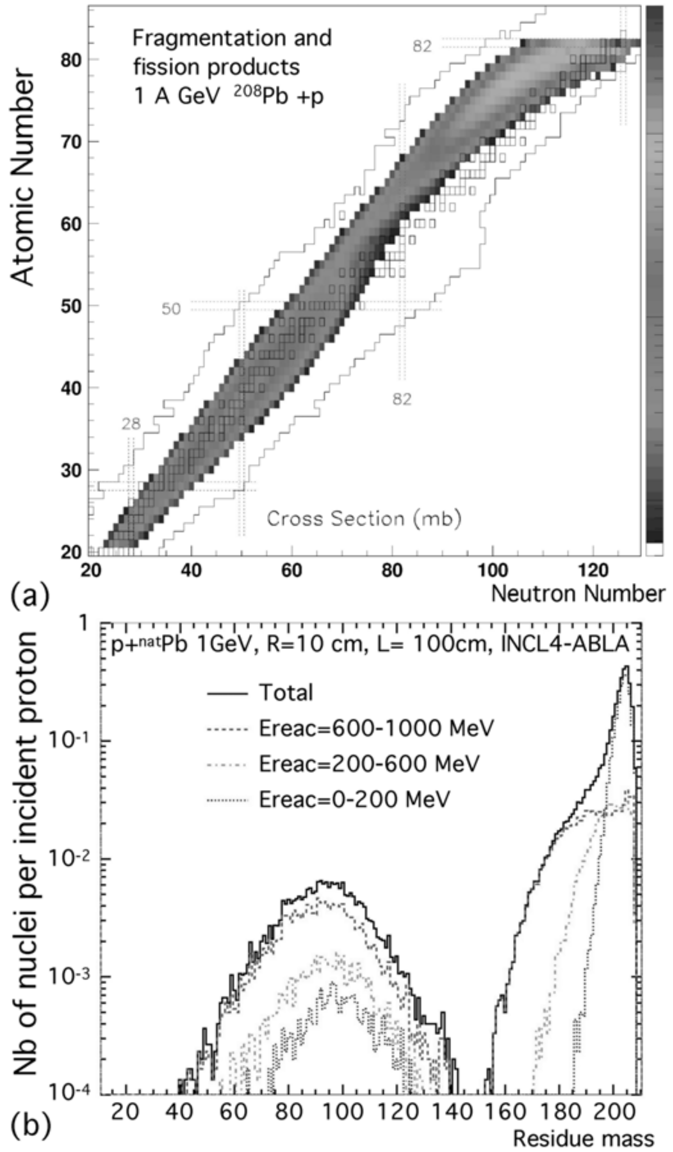

Figure 7. a) Nuclide production cross sections measured at GSI for the reaction p+Pb at 1 A GeV on a chart of the nuclides (Wlazlo et al. 2000, Enqvist et al. 2001), b) Number of nuclides per incident proton produced in a thick Pb target as a function of their mass for different bins in energy of the interaction.

The cross-sections of the thou-sand measured isotopes are represented on a nuclear chart, the empty squares indicating the stable nuclei. It can be seen that most of the residues are radioactive. Two main regions can be identified: one corresponding to evaporation residues beginning close to lead, which are mostly proton-rich nuclei, the other one due to fission residues, which represent about 10% of the total cross-section. In a thick target, a lot of residues are also produced by the secondary reactions. Actually, 3.5 isotopes per incident proton are created in average in a Pb target bombarded by a 1 GeV beam. The Figure 7b shows the mass distribution of the residues produced in the different interactions, divided into several bins in energy of the reaction, occurring in a thick lead target. It can be observed that the heaviest residues are created mostly in secondary low-energy (between 20 and 200 MeV) interactions while the fission products originate essentially from high-energy primary ones.

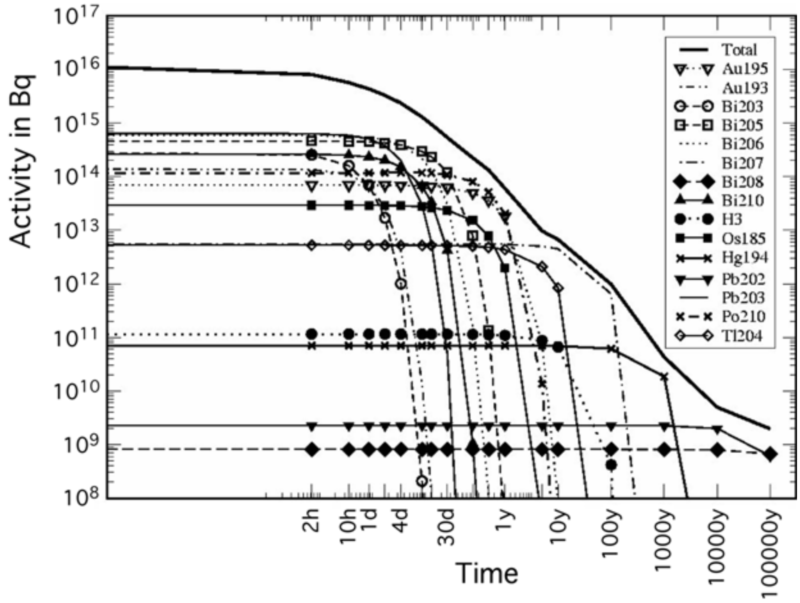

Figure 8. Main contributors to the activity calculated in the case of the liquid lead-bismuth MEGAPIE target. From Lemaire et al. (2007).

An important concern about spallation targets is the high activity generated by the numerous radioactive products. Figure 8 shows the total activity and the main contributors in the case of the MEGAPIE target (Lemaire et al. 2007). As expected from Figure 7b, the isotopes contributing the most to the activity are radioactive nuclides close to the target elements (Pb and Bi here). These nuclides are formed in spallation reactions in which only little excitation energy is deposited into the target nucleus, both because the energy of the interaction is low and because the highest cross-sections are due to peripheral collisions. Generally, the models used to simulate the reaction are well established for small energy transfers. Therefore, their predictions for these isotopes are very similar and reliable in a wide range of incident energies as shown in Leray 2003 in comparisons to measurements (Gloris et al. 2001) of excitation functions.

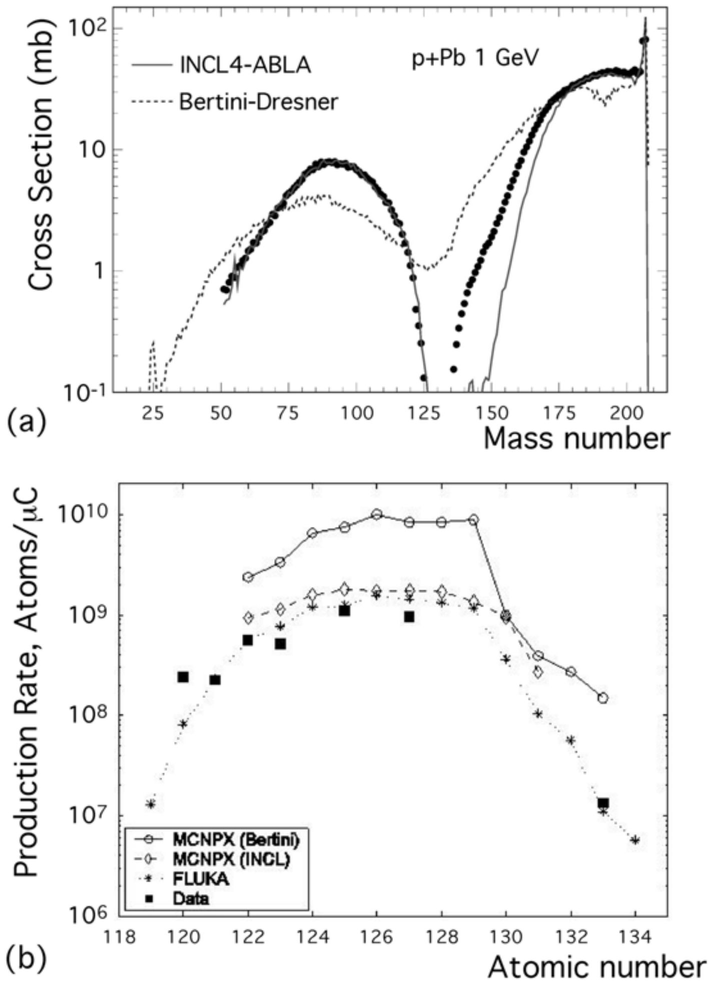

Figure 9. a) Comparison of INCL4-ABLA and Bertini-Dresner predictions with the production cross-sections as a function of the residue masses measured at GSI for the p+Pb system, b) Production rates of Xe isotopes from a thick Pb-Bi target, bombarded with 1.4 GeV protons, measured at Isolde, compared to different calculation. From Zanini et al. (2005).

Fission products are also important for radioprotection since some of them are gaseous elements, as Kr, Xe or I, which are released from the liquid metal during operation. Comparison of models to GSI elementary data have shown that Bertini-Dresner is not able to predict properly the fission residues (see Figure 9a) while INCL4-ABLA gives a very good agreement. The same is true in a thick target as shown in Figure 9b, in which the production rates of Xe from a Pb-Bi target irradiated by 1400 MeV protons measured at Isolde are compared to different calculations. INCL4-ABLA, but also FLUKA (Fasso et al. 2005), gives a reasonable agreement with the data while Bertini-Dresner overestimates the production rates by nearly one order of magnitude. This indicates that Bertini-Dresner should never be used to calculate fission residues.

Figure 9a also shows that a systematic misprediction of light evaporation residues is observed with INCL4-ABLA and other standard models (Boudard et al. 2002, Mashnik et al. 2005). This is even more pronounced in light systems as iron, not shown here, measured by Villagrasaet al. (2007) at GSI at different energies. This means that production rates of isotopes far from the target nucleus cannot be predicted with a good precision. This can be important for the assessment of some impurities in a given material. In addition, very light fragments with charge between 3 and 10, often called intermediate mass fragments, observed in a lot of experiments (Gloris et al. 2001, Herbach et al. 2006, Napolitani et al. 2004) are generally not predicted by the models (Leray 2003). Among those are for instance ^{7}Be and ^{10}Be, which are a concern for radioprotection. In the case of iron, a recent experiment (SPALADIN) has been performed at GSI, in which all the fragments and most of the particles emitted simultaneously in the reaction have been measured. This has allowed investigating in details the de-excitation mechanism and shown that intermediate mass fragments are probably formed in an asymmetric break-up mechanism (Le Gentil 2008), which is presently not taken into account in standard de-excitation models. The new version of ABLA is expected to solve this deficiency.

4. Conclusions

Spallation reaction in ADS targets lead to specific radioprotection and material damage problems. The predicting capabilities of nuclear models implemented into widely used high-energy transport codes can now be assessed, thanks to their validation on elementary data. It can be concluded that neutron production, heavy spallation residues and total activity of the spallation target can be calculated reliably by most of the models. As regards volatile fission element production, INCL4-ABLA or FLUKA should be preferred to the default option of MCNPX. For gas production and intermediate mass fragments, none of the available models are satisfying. Recent developments not yet implemented into transport codes are expected to cure these deficiencies.

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