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Energy multiplication and fissile fuel breeding limits of accelerator-driven systems with uranium and thorium targets

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ABSTRACT

The study analyses the integral ^{233}U and ^{239}Pu breeding rates, neutron multiplication ratio through (n,xn)- and fission-reactions, heat release, energy multiplication and consequently the energy gain factor in infinite size thorium and uranium as breeder material in an accelerator driven systems (ADS), irradiated by a 1-GeV proton source. Energy gain factor has been calculated as $M_{energy} = 1.67$, 4.03 and 5.45 for thorium, depleted uranium (100% ^{238}U) and natural uranium, respectively, where the infinite criticality values are $k_\infty = 0.40$, 0.752 and 0.816. Fissile fuel material production is calculated as 53 ^{232}Th(n,γ)^{233}U, 80.24 and 90.65 ^{238}U(n,γ)^{239}Pu atoms per incident proton, respectively.

The neutron spectrum maximum is by ~1 MeV. Lower energy neutrons $E < 1$ MeV have major contribution on fissile fuel material breeding (>97.5%), whereas their share on energy multiplication is negligible (0.2%) for thorium, depleted uranium. Major fission events occur in the energy interval 1MeV $< E <$ 50 MeV.

Introduction

Accelerator-driven systems

By the bombardment of heavy nuclei with $A > 200$ with high energetic protons, the so called spallation target will be destroyed and a great number of fast neutrons will be evaporated. Accelerator-driven systems (ADSs) couple the technology of high-intensity particle accelerator with spallation targets, which provide a great number of high energetic neutrons for a multiplicative sub-critical blanket/core. For ADSs, typical proton energies range from ~400 MeV up to 1.5 GeV and the tails of spallation neutron energies range from thermal up to GeV with maxima by 1–2 MeV. Typical target materials are lead, bismuth, thorium and uranium. The heavier the target nucleus and the higher the incident proton energy is the greater number of neutrons will emanate from the target nucleus so that thorium and uranium targets will generate more neutrons than lead and bismuth. On the other hand the latter can serve also as metallic coolant materials. Therefore, they are preferred target materials. The threshold

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fission energy for ^{232}Th and ^{238}U are ~1.4 MeV and ~0.8 MeV so that these breeder materials can undergo fission reactions at high neutron energies, creating secondary neutrons so that ADS will provide a neutron rich environment around the target material for an energy multiplying/fissile fuel material breeding blanket as well as for nuclear waste material transmutation. This has been recognized already in late 70's/early 80's, and a great number of technical papers have emerged in scientific literature [1–5]. Mainline concepts have proposed a 1 GeV/300 mA accelerator with a proton beam power of P_{acc} = 300 MW and a blanket multiplication factor or the so called gain factor G = ~10 for a 3000 MW power plant [1–5].

During the last 2 decades, a great number of works on ADS studies have emerged in the scientific literatures, which were all concerned with specific design concepts. Though proton beam energy in the range of ~1 GeV is now more or less conservative, proton beam currents of ten's or hundred's of mA range will require major technological progress and severe technological problems to overcome. Recent papers suggest bypassing this drawback by increasing the sub-criticality close to the critical value of $k_{eff} = 1.0$, in extreme limits up to 1–3 %. The famous Nobel Laureate Carlo Rubia is the main advocate of that line [6,7]. In a recent paper, he drives the sub-criticality even to $k_{eff} = 0.997$ and a gain factor of G = 700 [8]. The most advanced experimental ADS project MYRRHA [9] considers $k_{eff} = 0.97$ as highest permissible blanket criticality limit with respect to reactor safety [10]. Encouraged by Nobel Laureate's idea, a great number of papers with k_{eff} close to ninety have emerged. Kamil Tuček assumes $k_{eff} \leq 0.95$ in his PhD work at KTH for a blanket fueled with transuranium (TRU) nuclear waste material [11]. Per Seltborg calculates required proton beam current as i_p = 47, 22, 9 mA, proton beam power as P_{acc} = 47, 22, 9 and energy gain as G = 21, 45, 116 for a 1000 MW$_{th}$ core E_p = 1000 MeV in his PhD work at the same Institute [12]. In these early phases, the MUSE Experiments in the MASURCA Facility have been conducted with a core configurations of around $k_{eff} = 0.995$. In later phases, deeper sub criticality ($k_{eff} = 0.96$) was achieved by inserting a safety rod [13]. The SAD experiments have been conducted in order to investigate the basic physical principles of an ADS and thereby validating the theoretical predictions and estimations of the technological features of such systems. In the course of experiments, multiplication coefficient has been chosen in the window $k_{eff} = 0.95–0.98$ [14]. Preliminary Design Studies of an Experimental Accelerator Driven System (PDS-XADS) being supported by the European Commission are focused on options employing molten Lead–Bismuth Eutectic (LBE) and helium gas coolants, with a sub criticality of $k_{eff} = 0.96802$ and energy multiplication of M = 29.9 for LBE cooled blanket and $k_{eff} = 0.94877$ and M = 22.4 for gas-cooled blanket [15]. Neutronic properties of lead–bismuth-cooled accelerator-driven systems with different minor-actinide-based ceramic fuels (two composite oxides and one solid-solution nitride) have been investigated in Ref. [16]. The ADS model has used minor actinide-based fuels with relative fractions of the actinides Pu/Am/Cm = 0.4/0.5/0.1 for the helium cooled model in order to obtain a k_{eff} close to 0.97 [16].

It is true that a proton driven subcritical ADS with $k_{eff} < 1$ would increase the neutron level, and consequently the fission energy level proportional to the inverse of $1/(1-k_{eff})$. While such high k_{eff} values could be attained in a sub-critical blanket fueled with TRU nuclear waste materials or enriched uranium, nuclear physics puts significantly lower limits for natural breeder materials. On the other hand, availability of high quality nuclear fuel would make the use of such a complicated machine like fusion hybrid or an ADS hybrid obsolete, because, the same fuel can be used much simpler in conventional nuclear reactors with well known technology.

Thorium and uranium as nuclear fuel breeder

In a blanket containing thorium breeder material, primary and secondary neutrons will be captured and high quality nuclear fuel material ^{233}U will be produced. Conventional nuclear reactors use ^{235}U as nuclear fuel material. At present, only ~1% of uranium reserves can be exploited as nuclear fuel material under consideration of plutonium recycle. World thorium reserves are estimated to be about three times more abundant than the natural uranium reserves. Almost unlimited source of energy would be available with widespread breeding in thorium, which would require major new developments. At present, few numbers of liquid metal fast breeder reactors (FBRs) are in operation. However, doubling time of an FBR turns out to be longer than it was estimated at design stage. Hence, external neutrons are needed for the widely exploitation of nuclear fuel resources. There are two emerging technologies which can provide high energetic intense neutron source:

- Accelerator-driven systems,
- Fusion-fission (hybrid) reactors.

The latter has been widely studied in scientific literature [17–29]. Main characteristics of a prospective thorium energy scenario can be cited as follows:

- ❖ Two neutrons are necessary to initiate fission in critical reactors, $\eta > 2$.
- ❖ Only natural material thorium will be consumed. Enrichment is no longer necessary.
- ❖ Energy generation will be multi hundreds times larger than the one currently available from using only the ^{235}U fuel.

Fig. 1 and Table 1 compare the nuclear quality of main nuclear fuel materials ^{233}U, ^{233}U and ^{239}Pu in terms of neutron production η per neutron capture in the fuel as a function of neutron energy. ^{233}U has the smallest parasitic capture cross section and capture/fission ratio, both for fast and thermal reactors. One can clearly see that ^{233}U would be the best nuclear fuel for thermal breeders and ^{239}Pu for fast breeders. On the other hand, ^{235}U has highest delay neutron fraction β_{eff}, which offers a greater advantage for reactor control.

Thorium cycle produces ^{233}U which, from a non-proliferation point of view, is preferable to plutonium for two reasons. Firstly, it is strongly contaminated with ^{232}U, which decays to give highly radio-active daughter products. This leads already to a high level of deterrence and would make handling and diversion difficult, even impossible for clandestine misuse by terrorist groups or states. Secondly, the ^{233}U could easily be denatured with ^{238}U by adding few fractions of natural uranium to thorium. The thorium option not