Energy Production and Transmutation of Nuclear Waste by Accelerator Driven Systems

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Abstract. There is a significant amount of highly radiotoxic long-life nuclear waste (NW) produced by NPP (Nuclear Power Plants). Transmutation is a process which transforms NW into less radiotoxic nuclides with a shorter period of half-life by spallation neutrons or radiative capture of neutrons produced by ADS (Accelerator Driven System). In the processes of transmutation new radioactive nuclides are produced. ADS is big energy consumer equipment. It is a method for production of a high-flux and high-energy neutron field. All these processes occur in ADS simultaneously. ADS is able to transmute actinides and produce energy simultaneously. The article considers the energy production problems in ADS. Several ideas are developed regarding the solution of the global energy supply.

1. Introduction

In the mid-90s many experiments and theoretical investigations of the parameters and possibilities of ADS (Accelerator Driven System) were started [1, 2, 3, 4, 5, 6]. The investigations may be divided into two parts, experimental and computer simulations. Numerous experiments for determination of the high-energy cross sections and estimation of the high-energy neutron field inside and outside the experimental set-ups have been done. Computer simulations with MCNPX, GEANT4, FLUKA transport codes were used. Most of the codes use group cross sections for calculations, whereas MCNPX uses another method of precise nuclear data tables which are included in the program. Most of the nuclear data tables are defined up to 20 MeV. Some of the tables are extended up to 100 or 150 even up to 200 MeV [7]. The group cross sections do not describe accurately the neutron induced reactions for neutrons with energy $E_n<1$ MeV because of resonances. The MCNPX transport code is the most powerful code describing nuclear fission. All codes use nuclear models if there are no data tables. MCNPX is the most advanced transport code for calculation of ADS integral energy parameters (described below) with significant amount of fission nuclides such as $^{239,240}$Pu and $^{233,235}$U. The new cyclotron centre based on TR24 cyclotron at the Institute for Nuclear Research and Nuclear Energy of the Bulgarian Academy of Sciences opens new possibilities for experimental research in the field of accelerator based neutron sources for applied research and nuclear medicine [8].

2. ADS design

There are many suggestions concerning the ADS design. However, first of all the main aims of ADS should be defined. The article aims to investigate the energy production by ADS. The energy is produced by neutron induced fission in isotopes $^{233,235,238}$U, $^{232}$Th and $^{239,240}$Pu. All ADS calculations are made by the MCNPX transport code (Bertini model) and (Evaluated Nuclear Data File) ENDF60 data table [9]. The design of ADS is simple because we are interested only in integral parameters such as total neutron production, leakage of neutrons and neutron induced reactions. In general, ADS consists
of 4 main parts. The first is the accelerator. A proton beam with energies from 0.66 to 2 GeV is chosen. The second part is a target zone made of $^{nat}U$, $^{nat}Pb$ and a combination of $^{nat}Pb$ and W. The third part is a reflector and a cooling system, both made of $^{nat}Pb$. The forth is nuclear fuel MOX (Mixed Oxide Fuel, 20-30% concentration of isotopes $^{239}Pu+^{235}U$). For axial (direction of proton beam) symmetry the ADS has cylindrical shape with approximate dimensions: diameter $D=2.5$ m, length $L=2.5$ m. The approximate masses of nuclides without shielding ($^{nat}U$ and $^{232}Th$) are $M^{nat}U=270$ kg, $M^{239}Pu=109$ kg, $M^{238}Pb=2$ kg, $M^{nat}Pb=4.5$ t. The masses of the ADS shielding are approximately 100 t and 63 t of $^{nat}U$ and $^{232}Th$, respectively. The effective multiplication factor $K_{eff}$ is 0.98-0.99 for all cases.

![Figure 1. Cross sections of ADS. Target zone ($^{nat}Pb$, $^{nat}U$ or W), MOX – fuel, reflector $^{nat}Pb$ and $^{nat}U$ or Th protection shield (area) or (nuclear fuel production zone). $K_{eff} = 0.98 – 0.99$. The size is approximately 2.5x2.5 m.](image)

![Figure 1a. Axial cross section of ADS, cases 2 and 3.](image)

![Figure 1b. Cross section perpendicular to the axis of cylinder, cases 2 and 3.](image)

### 3. ADS simulations

Three cases are chosen for the simulations. The first case is an ADS system without shielding of $^{nat}U$ or $^{232}Th$, but it has a biological shielding and a steel holder of ADS. The biological shielding and the steel holder do not change the neutron field inside ADS, it is important only for the neutron leakage. Three different targets and proton beams with energies $E_{p}=0.66$, 1 and 2 GeV (the results are shown in tables 1 and 2) are included in the calculations. The second case is an ADS with $^{nat}U$ shielding and proton beam $E_{p}=1$ GeV and the third case is an ADS with $^{232}Th$ shielding and proton beam $E_{p}=1$ GeV (the results are shown in table 3). For all cases the number of $(n,f)$ reactions for incident neutrons with energies $E_{n}<20$MeV is calculated. High energy neutrons and charged particles induced fissions are not included in the calculations. The mass of the lead reflector in cases 2 and 3 is insignificantly higher than that in case 1. The steel holder and the biological shielding (in case 1) are removed and replaced by lead (in cases 2 and 3). The capture cross section of the iron nucleus is high, and because of that, the iron is not included in cases 2 and 3. The aim of $^{nat}U$ and $^{232}Th$ shielding is nuclear fuel production by reactions:

$$^{238}U(n,\gamma) \rightarrow ^{239}U \rightarrow ^{239}Np \rightarrow ^{239}Pu$$

$$^{232}Th(n,\gamma) \rightarrow ^{233}Th \rightarrow ^{233}Np \rightarrow ^{233}U$$

The neutron leakage is significant in case 1 and the neutrons will participate in unwanted capture reactions. The best way to avoid this is to place the $^{nat}U$ and $^{232}Th$ shielding. For ease of calculations the fission energy per one split is set $E_{n}=200$ MeV. This approximation will not change the integral results. In the beginning of the simulations, the total ionization energy deposition of the proton beam in the matter is investigated. The targets ($^{nat}U$, $^{nat}Pb$ and $^{nat}P+W$) with dimension diameter $D=5$ cm and length
L=100 cm are irradiated by protons with different energies. The ionization energy deposition $E_{\text{ion}}$ for every millimetre in the targets is calculated. The ionization may be calculated with the Bethe equation.

$$-\frac{dE}{dx} = \frac{4\pi n z^2}{m_e \beta^2} \left( \frac{e^2}{4\pi \varepsilon_0} \right)^2 \left[ \ln \left( \frac{2m_e e^2 \beta^2}{\mu (1-\beta^2)} \right) - \beta^2 \right]$$

where $n = \frac{N_A Z \rho}{\mu}$, $m_e$ – electron mass in rest, $c$ – speed light in vacuum, $\beta=v/c$, $v$ – speed of the particle, $\varepsilon_0$ – vacuum permittivity, $N_A$ – Avogadro number, $Z$ – charge of the medium, $\rho$ - medium density, $A$ – atom mass of medium, $\mu$ – molar mass of the medium. The Bragg-peak of protons is presented (figure 2).

The next step is $K_{\text{eff}}$ calculations. In the real system of energy production the equilibrium concentration of fission nuclides is supposed to be achieved. It means that the number of fissions reactions is less than the number of radiative capture of neutrons by fission nuclides, i.e. the rate of production of nuclear fuel is higher than the rate of consumption of nuclear fuel.

Table 1. Neutron balance in ADS (without $^{235}$U or $^{232}$Th shield) for a proton beam with energies $E_p=0.66, 1$ and $2$ GeV and three different targets $^{235}$U, $^{208}$Pb and $^{208}$Pb/W with dimensions $D=5$ cm and $L=50$ cm. $N_{\text{total}}$ is the total neutron production, $N_{\text{escape}}$ is the leakage of neutrons, $(n,f)$ and $(n,\gamma)$ are the number of reactions and “/” separation of the reactions. The calculations are made for one proton. $K_{\text{eff}} = 0.98 – 0.99$

| $E_p$ GeV | $N_{\text{total}}$ | $N_{\text{escape}}$ | $(n,f)^{235}$U | $(n,f)$, $(n,\gamma)^{239}$U | $(n,f)$, $(n,\gamma)^{239}$Pu | $(n,\gamma)^{240}$Pu |
|-----------|-------------------|-------------------|-----------------|----------------------|---------------------|------------------|
| 0.66      | 1350              | 363               | 6               | 37                   | 106                 | 330              |
| 1         | 2630              | 706               | 12              | 70                   | 190                 | 636              |
| 2         | 5540              | 1490              | 24              | 140                  | 407                 | 1364             | 255 | 23 |
I shows that this ADS construction cannot achieve an equilibrium stage despite the fact that $K_{\text{eff}}$ is less than 1, for example approximately 0.98-0.99. For all simulations $K_{\text{eff}}$ ranges between 0.985 and 0.998. To compare different experimental and computer simulations Beam Power Gain (BPG) is introduced. It is the ratio of the total energy production [MeV] and the beam energy [MeV]. The total energy production is presented by the equation: $E_{\text{tot}}=E_{\text{fiss}}+E_{\text{ion}}+E(n,\gamma)$, where $E_{\text{fiss}}$ is energy received by fission, $E_{\text{ion}}$ is ionization energy deposition of all ions and $E(n,\gamma)$ is the energy which is produced by the fission of the nuclides $^{233}\text{U}$ and $^{239}\text{Pu}$, received by reactions (1). In table 2, the beam power gain is calculated by $BPG=(E_{\text{fiss}}+E_{\text{ion}})/E_{p}$.

In that equilibrium stage, the concentrations and distribution of nuclides (fission and reflector) should be such that the $K_{\text{eff}}$ is less than 1, for example approximately 0.98-0.99. For all simulations $K_{\text{eff}}$ ranges between 0.985 and 0.998. To compare different experimental and computer simulations Beam Power Gain (BPG) is introduced. It is the ratio of the total energy production [MeV] and the beam energy [MeV]. The total energy production is presented by the equation: $E_{\text{tot}}=E_{\text{fiss}}+E_{\text{ion}}+E(n,\gamma)$, where $E_{\text{fiss}}$ is energy received by fission, $E_{\text{ion}}$ is ionization energy deposition of all ions and $E(n,\gamma)$ is the energy which is produced by the fission of the nuclides $^{233}\text{U}$ and $^{239}\text{Pu}$, received by reactions (1). In table 2, the beam power gain is calculated by $BPG=(E_{\text{fiss}}+E_{\text{ion}})/E_{p}$.

Thus, shielding made of $^{nat}\text{U}$ or $^{232}\text{Th}$ is placed to increase nuclear fuel production. The results with two different types of shielding are presented in table 3.

### Table 2. Fission $E_{\text{fiss}}$ [x10^4MeV] and ionization $E_{\text{ion}}$ [x10^4MeV] energy deposition in ADS for three targets $^{nat}\text{U}$, $^{nat}\text{Pb}$ and $^{nat}\text{P+W}$ and proton beam energies $E_{p}=0.66$, 1 and 2 GeV are presented. The ratio $E_{\text{tot}}/E_{p}$ shows the energy amplifier of the ADS without $^{nat}\text{U}$ or $^{232}\text{Th}$ shield. The calculations are done for one proton. $K_{\text{eff}}=0.98-0.99$.

| target   | $E_{p}=0.66\text{GeV}$ | $E_{p}=1\text{GeV}$ | $E_{p}=2\text{GeV}$ |
|----------|-----------------------|---------------------|---------------------|
| $^{nat}\text{U}$ | $E_{\text{fiss}}$ | $E_{\text{ion}}$ | $E_{\text{tot}}/E_{p}$ | $E_{\text{fiss}}$ | $E_{\text{ion}}$ | $E_{\text{tot}}/E_{p}$ | $E_{\text{fiss}}$ | $E_{\text{ion}}$ | $E_{\text{tot}}/E_{p}$ |
| $^{nat}\text{Pb}$ | 9.0 | 0.65 | 147 | 15.5 | 1.1 | 166 | 35.5 | 2.5 | 190 |
| $^{nat}\text{P+W}$ | 6.8 | 0.5 | 110 | 12.8 | 0.92 | 127 | 29.2 | 2.5 | 158 |

Thus, shielding made of $^{nat}\text{U}$ or $^{232}\text{Th}$ is placed to increase nuclear fuel production. The results with two different types of shielding are presented in table 3.

### Table 3. Neutron induced reactions (n,f) and (n,\gamma) in ADS (with $^{nat}\text{U}$ or $^{232}\text{Th}$ shield) for proton beam with energy $E_{p}=1\text{GeV}$. The neutron leakage is 2.5% and 5% for $^{nat}\text{U}$ or $^{232}\text{Th}$ shield, respectively. The calculations are made for one proton. $K_{\text{eff}}=0.98-0.99$.

| $^{nat}\text{U}$ - shield | (n,f)$^{235}\text{U}$ | (n,f), $^{(n,\gamma)}^{238}\text{U}$ | (n,f), $^{(n,\gamma)}^{239}\text{Pu}$ | (n,f), $^{(n,\gamma)}^{240}\text{Pu}$ |
|---------------------------|---------------------|---------------------|---------------------|---------------------|
| MOX                       | 100                 | 144                 | 430                 | 1410                | 272                | 6                  |
| Target                    | 2                   | 7                   | 30                  | -                   | -                  | -                  |
| Shield                    | 140                 | 55                  | 1960                | -                   | -                  | -                  |

| $^{232}\text{Th}$ - shield | (n,f)$^{235}\text{U}$ | (n,f), $^{(n,\gamma)}^{238}\text{U}$ | (n,f), $^{(n,\gamma)}^{239}\text{Pu}$ | (n,f), $^{(n,\gamma)}^{240}\text{Pu}$ |
|---------------------------|---------------------|---------------------|---------------------|---------------------|
| MOX                       | 75                  | 105                 | 310                 | 1020                | 200                | 5                  |
| Target                    | 2                   | 6                   | 25                  | -                   | -                  | -                  |
| Shield                    | 0                   | 1220                | -                   | -                   | -                  | -                  |

The comparison of nuclear fuel production by reaction (n,\gamma)$^{238}\text{U}$ and fission reaction (n,f)$^{239}\text{Pu}$ in table 1 shows that this ADS construction cannot achieve an equilibrium stage despite the fact that $K_{\text{eff}}$
=0.99. The fissions are approximately three times more than neutron capture reactions (nuclear fuel production). The neutron leakage is significant.

Table 4 shows the integral number of (n,f) reactions and the number of production of nuclear fuel $^{239}$Pu, $^{233}$U by radiative capture and BPG. The equilibrium stage is reached. Moreover, the production rate of nuclear fuel is higher than the fission rate. Most of the new nuclear fuel is generated in the shielding, not in the MOX or target areas. This is good, because the new nuclear fuel can be removed from ADS during irradiation.

### Table 4. The total neutron induced fission, production of $^{239}$Pu and $^{233}$U are shown. The energy amplifier BPGs are presented in the last two columns. The proton beam energy is $E_p=1$ GeV. E(n,$\gamma$) is the fission energy of produced nuclear fuel $^{239}$Pu and $^{233}$U. $K_{\text{eff}}=0.99$. The calculations are made for one proton.

| Shield | (n,f)$_{\text{total}}$ | (n,$\gamma$)$_{^{238}\text{U}}$ | (n,$\gamma$)$_{^{232}\text{Th}}$ | BPG=E$_{\text{fiss}}$/E$_p$ | BPG=(E$_{\text{fiss}}$+E(n,$\gamma$))/E$_p$ |
|--------|----------------|----------------|----------------|----------------|----------------|
| natU   | 1850           | 2400           | 370            | 850            |
| $^{232}\text{Th}$ | 1200          | 310            | 240            | 540            |

Most of the fissions come from (n,f)$^{239}$Pu reaction and the fission cross sections for low energy neutrons are very high. Part of these neutrons are obtained after the interaction with the neutron moderator $^{nat}$Pb. Therefore, the additional simulations are done with different data libraries for $^{nat}$Pb, the same cross sections for other fission nuclides and the same high energy nuclear model. The total number of fissions in ADS calculated by MCNP6 with ENDF60 data tables [9] (for $^{nat}$Pb) are less than that of the fissions calculated with ENDF60. The total number of fissions calculated by MCNP6 and RMCCS or DRMCCS data tables (for $^{nat}$Pb) are higher than that of the fissions calculated with ENDF60. Similar behaviour is observed in fuel production in such ADS structure. For ADS calculations the cross sections data tables are very important as well as nuclear models.

### 4. Discussion

The ADS is supposed to be controlled by the parameters of the proton beam, but the significant presence of MOX ($^{239}$Pu) in the reactor part requires more calculations and investigations. A sudden increase of the $^{239}$Pu fissions to $K_{\text{eff}}>1$ is possible, which is out of the control of the accelerating proton beam. The neutron field in ADS is highly inhomogeneous. Close to the target the neutron field has many spallation neutrons. The neutron field in the reactor part (MOX – fuel) consists of fission neutrons, spallation neutrons and reflected neutrons from the lead. The neutron field in the $^{nat}$U or/and $^{232}$Th shield (nuclear fuel production region) consists of fission and neutrons reflected from the lead. Energy production strongly depends on the Pb cross section neutron moderation. Moreover, the $^{206,207}$Pb isotopes can capture low energy neutrons during irradiation and the Pb isotope concentration in the moderator will change.

### 5. Conclusion

In the reactor part no equilibrium (production rate of nuclear fuel higher than fission rate) can be reached. The equilibrium can be achieved if additional layers of $^{dep}$U or $^{232}$Th are placed around ADS (case 2 and 3). These layers produce nuclear fuel and reduce the neutron leakage to the minimum.

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