The Progress of Researches on ADS in China

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Abstract. The conceptual study of the accelerator driven system (ADS) which lasted for about five years ended in 1999 in China. As one project of the National Basic Research Program of China (973 Program) in energy domain, which is sponsored by the China Ministry of Science and Technology (MOST), a five-year-program of fundamental research of ADS physics and related technology was launched in 2000 and passed national review at the end of 2005. From 2007, another five-year 973 Program, Key Technology Research of Accelerator Driven Sub-critical System for Nuclear waste Transmutation, started. The research activities will focus on HPPA physics and technology, reactor physics of external source driven sub-critical assembly, nuclear data base and material study. For HPPA, a high current injector consisting of an ECR ion source, LEBT and an RFQ accelerating structure of 3.5 MeV has been built and will be improved. In reactor physics study, a series of neutron multiplication experimental study has been carried out and still being done. The VENUS facility has been constructed as the basic experimental platform for neutronics study in ADS blanket. It's a zero power sub-critical neutron multiplying assembly driven by external neutron produced by a pulsed neutron generator. The theoretical, experimental and simulation study on nuclear data, material properties and nuclear fuel circulation related to ADS is carrying on to provide the database for ADS system analysis.

1. Introduction

China, as a developing country with a great population and relatively less energy resources, actively emphasizes the development of nuclear energy. To develop nuclear power in such a large scale, two problems must be solved. First, as technically and economically exploitable natural uranium resources are limited domestically or overseas, uranium utilization rate has to be greatly raised. Second, long-lived radioactive nuclear wastes have to be safely disposed to reduce the impact on the environment and to eliminate public fear of nuclear power.

Right now only a small amount of spent fuels from NPPs has been accumulated in China. But the situation will be very serious in the future according to the prediction of nuclear energy development in China. The annual generated waste is estimated to be 7 500 m^3 and 10 000 m^3 respectively for the year 2010 and 2020.

Considering MA and LLFP transmutation with more efficiency and non-criticality risk for new nuclear application the accelerator-driven sub-critical system (ADS) have been started to develop as a national research projects in China.

The conceptual study of the accelerator driven system (ADS) which lasted for about five years ended in 1999 in China. As one project of the National Basic Research Program of China (973 Program) in energy domain, which is sponsored by the China Ministry of Science and Technology (MOST), a five-year-program of fundamental research of ADS physics and related technology was launched in 2000 and passed national review at the end of 2005[1,2,3]. From 2007, another five-year 973 Program, Key Technology Research of Accelerator Driven Sub-critical System for Nuclear waste Transmutation, started. China Institute of Atomic Energy (CIAE), Institute of High Energy Physics (IHEP), School of Nuclear Science and Engineering in Shanghai Jiao Tong University, Institute of Heavy Ion Physics in Peking University (PKU-IHIP) and other institutions jointly carried out the research. The research activities will focus on HPPA physics and technology, reactor physics of external source driven sub-critical assembly, nuclear data base and material study. For HPPA, a high current injector consisting of an ECR ion source, LEBT and an RFQ accelerating structure of 3.5 MeV has been built and will be improved. In reactor physics study, a series of neutron multiplication experimental study has been carried out and still being done. The VENUS facility has been constructed as the basic experimental platform for neutronics study in ADS blanket. It's a zero power sub-critical neutron multiplying assembly driven by external neutron produced by a pulsed neutron generator. The theoretical, experimental and simulation study on nuclear data, material properties and nuclear fuel circulation related to ADS is carrying on to provide the database for ADS system analysis.

In the last few years the scientific and technical exchange and cooperation with foreign research institutions in different aspects are of great help to the related work.

Some important results were summarized as follows.

2. Venus I experiment—The measurement on sub-critical assembly driven by pulsed external source

A composed structure of zero-power sub-critical assembly combined with a pulsed neutron source, Venus I program is currently being carried on. The pulse-neutron was provided by a Cockroft-Walton machine, routinely operated since 2001. 14 MeV and 2.5 MeV neutron was derived by d-T and d-D reaction. The neutron yield in DC mode can reach 10^{12}n/s, while in micropulse mode 10^{9}n/s~10^{10}n/s for d-T reaction.

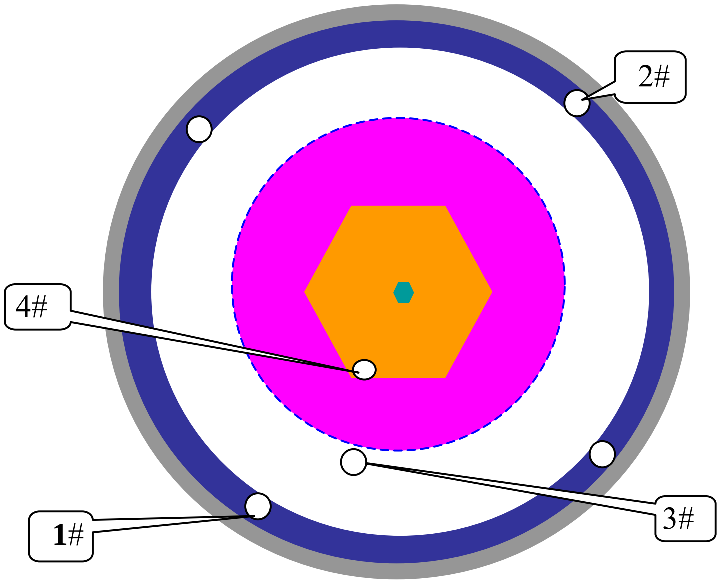

Fig.1 *The subcritical reactor core arrangement of the Venus I program*

As shown in Fig.1, there are source and buffer in the center of the composed core, a driven zone consisting of natural Uranium pin is very dense lattice with aluminum in between, an active zone with 20% and mainly 3% enriched ^{235}U fuel pin is polyethylene lattice and the polyethylene reflector. Different neutron spectra in different zone are expected. The buffer will shift the sharp 14 MeV and 2.5 MeV neutron to the fast neutron spectra to mock-up the evaporation bump in the spallation neutron spectrum and fission spectrum as possible as. In the driven zone not much neutron multiplication is expected, while the hard neutron spectra with an average energy about 700 keV is expected. In the active zone, thermal neutron is expected. The assembly will be operated in deep sub-criticality $k_{\text{eff}} \approx$ 0.90–0.95 range.

On July 18, 2005, the first fuel element was loaded into Venus-1 sub-critical assembly and the following preliminary experiments were subsequently conducted: determination of fuel loading, measurement of relative importance of external source at different positions, measurement of radial and axial thermal neutron flux distributions in the fast zone, and measurement of thermal neutron count changing with the fuel loading in the fast zone.

During the experiment, detectors were placed at the outer of polyethylene reflector and in the outer layers of the fast zone respectively. The detectors are one ^{3}He counter with dimensions of Φ10×150mm as 1# detector, two BF_3 counter of Φ65×780mm as 2# and 3# detectors and two gamma-compensated ionization chambers (Φ50×560mm).The ^{3}He and BF_3 proportional counters were used for inverse multiplication measurement (critical approach). Gamma-compensated ionization chambers used for power monitoring. An additional detector 4 # was put in the vertex of the hexagonal fast zone (in the tenth layer) for observing neutron count trend.

The results of the preliminary experiments show that the design objectives and requirements for China's ADS sub-critical assembly Venus-1 have been fulfilled[4]:

- The effective neutron multiplication factor k_{eff} of the coupled core with a fast zone and a thermal zone is variable within a range of 0.90-0.98, hence the assembly is suitable for the study on neutron behavior of sub-critical reactor driven by an external neutron source.
- The average neutron energy is higher than 600KeV at a distance 4~36cm from the centre of the core, with the highest neutron energy at a position near the neutron source zone. The design requirement of neutron energy is satisfied. The fast zone can be used for transmutation research.
- There is a suitable space for transmuting MA in the fast neutron zone and sufficient fission for energy generation in the thermal neutron zone. There is also a epithermal neutron zone between the fast neutron zone and the thermal neutron zone and can be used for LLFP transmutation.
- The neutron importance of an external source is largest in the core center, indicating that the target for producing spallation neutrons should be placed in the core center region.
- The relative distribution of thermal neutron flux in 1-8 layers of the fast zone is very low. Whether it means a higher fast neutron flux in this region suitable for transmuting MA needs to be verified in the future. The relative distribution of thermal neutron flux in 8–10 layers is high due to the effect of thermal neutrons in the thermal zone, suitable for transmuting LLFR. Thermal neutron flux at the 10th layer (–42.5cm) is high due to the effect of the end reflector in the thermal zone. The asymmetry of axis distribution of thermal neutron flux is attributed to the end reflector mounted only at one side.
- The neutron counts of detectors at different positions vary in different ways with the loading of natural uranium fuels into the fast zone. The neutron count of detector 4# first increases to a peak value during the loading of the 1st- 6th layer of natural uranium fuel, then decreases when loading the 7-10th layer of the fuel rods. It may be explained by the effect of neutron energy of the external neutron source, as the spectrum of Am-Be steady neutron source has four peak energy, i.e. 3.5MeV, 5.0MeV, 8.0MeV and 10.0MeV, all greater than fission threshold of natural uranium (1.1MeV) and inevitably causing fast fission of U-238. While the effect of fast fission caused by Am-Be source can reach the 1st-6th layer, it becomes small in the 7th-10th layer. It has been confirmed that the effective range of fast fission caused by external source will extend with the increase of external neutron source energy. This has been done using D-T reaction source whose neutron energy is 14MeV.

Recently some experiments are performed on Venus-1 using the Cf-252 and Am-Be source as external neutron source. The relative counting rate curve vs adding layer of fuel rods are shown in fig 2.

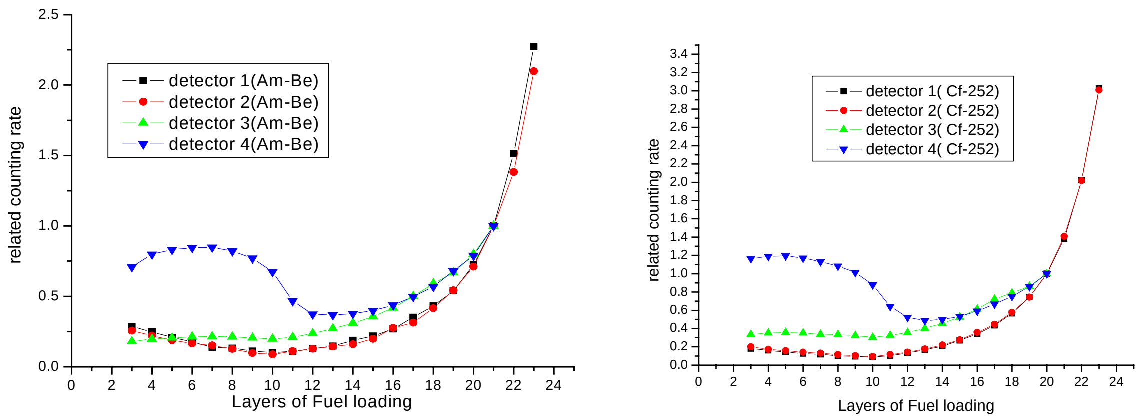

Fig 2 *The relative counting rate curve vs adding layer of fuel rods with Am-Be and Cf-252 source*

3. Neutronics and thermal-hydraulics technology research of ADS –Venus 2

A conceptual design of ADS verification facility-Venus Ⅱ has been performed. The main parameters are listed in table 1.

Table 1. The Main Parameters of Venus //

Fuel	Spent fuel of CARR, U_3Si_2-Al, 149.3kg
Keff	0.982
Spallation Target	Solid W
Energy of Proton Beam	100MeV
Yield of spallation neutron	0.3 n/p
Beam Intensity	0.3 mA
Beam Power	30 kW
Thermal Power of the Core	200kW

A primary design of a LBE loop for thermo-hydraulic study and material study has been performed. The main parameters are listed in table 2.

Table 2. The Primary Parameters of LBE loop

Highest Temperature	550 ℃
Maximum Flux	6 m^3/h （velocity 3 m/s）
Pressure	0.3 MPa
Oxygen Control	Ar＋5%H_2/H_2O
LBE capacity	100～150 l
Height of Loop	5 m
Experimental Segment	2
Height of Segment	1.5～2 m
Velocity of Flux	1 m/s
Temperature Difference	100 ℃

4. Intense proton ion source

An electron cyclotron resonance (ECR) ion source[5] is selected for the source of our verification facility system. The microwave power generated by a 2.45 GHz-1 kW magnetron is coupled into the copper chamber (54 mm ×72 mm in cross section and 36 mm long) through a three stub tuning unit and ridged wave guide. Inside the chamber, a ϕ54 mm in diameter quartz tube which is tightly fixed by a BN disk and plasma electrode is placed to confine the plasma. A BN plate is placed between the ridged waveguide and plasma chamber to separate the plasma and vacuum. Three holes are made on the waveguide to evacuate the wave guide after the microwave window; with this configuration the gas in the waveguide can be evacuated quickly to avoid interfering with the discharge. The microwave window for vacuum sealing is placed behind a bent section in order to avoid any damage due to the back streaming electrons. The microwave system including its power supply is placed on the 75 kV high voltage platform.

A 90 mA hydrogen beam can be routinely extracted from a ϕ6.5 aperture of the source. The emittance of the extracted beam is measured by multi-slits and single thread emittance-measuring unit. The measured emittance of the total beam at 90 mA, 60 kV, 50 cm downstream of the ion source is $0.129\pi\,\text{mm}\cdot\text{mrad}$. At a specific extraction distance, an adequate extraction voltage always can be found for various beam currents to obtain minimum emittance. The proton ratio is measured by analyzing a portion of the beam with a mini-deflection magnet. The result shows that proton fraction is more than 80% which satisfies the requirement of the system. The proton fraction slightly varies with the changes of microwave power but no significant effect is found.

5. RFQ accelerator study

The structure of RFQ is a four-vane type and designed to accelerate 50 mA peak current of proton beam with input energy of 80 kV. In the preliminary research phase, the 352.2 MHz RF system will be operated in pulse mode. CERN kindly provided IHEP with some RF equipment. Because the given RF system was used for CW operation at CERN before, to apply them to our pulse mode operation, some modifications and improvements are necessary. We have made some indispensable assemblies, and also did some tests and commissioning of every sub-system. For example, we have finished the 100 kV power supply test and long pulse floating desk hard tube modulator test. Furthermore, the initial high power condition of the klystron is carried out, and output power can reach up to 800 kW.

The fabrication of the RFQ copper model was done by a company in Shanghai, China. At first, some tests for development the mechanical technology had to be done, for example, the brazing technology for assembling four vanes together with required mechanical tolerance, the characteristics of melting filler, the structure surface and the vacuum leak; the drilling of the coolant hole through the 1.2 m RFQ cavity with 12 mm in diameter; the precision machining of the vane electrodes on the numerical controlled mill. The fabrication of the RFQ was finished last year.

After setup of the ion source, the LEBT and RFQ, we started RFQ beam commissioning on 7 July, 2006. We immediately got a beam of 20 mA from RFQ with a transmission rate of about 60%. After a few days, a 41 mA beam comes from the RFQ and the transmission rate reaches 92%. The duty factor is 15% at the beginning of 2009.

6. ADS related nuclear data

The new nuclear reaction theoretical models code MEND, which can give all kinds of reaction cross sections and energy spectra for six outgoing light particles (neutron, proton, alpha, deuteron, triton, and helium), gamma and recoil nuclei in the energy range up to 250 MeV, is being developed. The incident particle can be neutron, proton, alpha, deuteron, triton and helium. A program [6] for automatically searching optimal optical potential parameters in E <300 MeV energy region has been developed. By this code the best optical potential parameters can be searched automatically to fit with relevant experimental data of total cross sections, nonelastic scattering cross sections, elastic scattering cross sections and elastic scattering angular distributions. The nuclear data evaluation method has been developed for ADS. According to the experimental data of neutron-induced reactions, and theoretical model calculation codes UNF[7], ECIS and DWUCK, all cross sections of neutron induced reaction, angular distributions, double differential cross sections for neutron, proton, deuteron, triton, helium and alpha emission, γ -ray production cross sections and γ -ray production energy spectrum are calculated and evaluated at incident neutron energies from 10^{-5} eV to 20 MeV. Since the recoil effect is taken into account, the energy for whole reaction processes is balance. Nuclei have been evaluated as follow: ^{28,29,30}Si, ^{50,52,53,54,nat}Cr, ^{54,56,57,58,nat}Fe, ^{90,91,92,94,96,nat}Zr, ^{93}Nb, ^{112,114,115,116,117,118,119,120,122,124,nat}Sn, ^{180,182,183,184,186,nat}W[8], ^{204,206,207,208,nat}Pb[9], ^{209}Bi, ^{232}Th, ^{233,234,235,238}U[10] etc. By using advanced nuclear models that account for details of nuclear structure and the quantum nature of the nuclear scattering. The nuclear data are calculated and evaluated for both incident neutrons and incident protons at incident neutron energy from 20 to 250 MeV as follow: ^{27}Al, ^{30}Si, ^{50,52,53,54}Cr, ^{54,56,57,58}Fe, ^{90,91,92,94,96}Zr, ^{180,182,183,184,186}W, ^{181}Ta, ^{204,206,207,208}Pb, and at incident proton energy from threshold energy to 250 MeV as follow : ^{54,56,57,58}Fe[11], ^{180,182,183,184,186}W, ^{204,206,207,208}Pb, ^{209}Bi.

7. ADS related target physics

The calculations for the standard thick target were made using different codes. The simulation of the thick Pb target with a length of 60 cm, diameter of 20 cm bombarded with 800, 1000, 1500, and 2000 MeV energetic proton beam was carried out. The yields and spectra of emitted neutron were studied. The spallation target was simulated by SNSP, SHIELD, DCM\CEM (Dubna Cascade Model \Cascade Evaporation Mode), and LAHET codes[12,13]. The neutron yields calculated by SHIELD and DCM\CEM were in agreement within ±10%.

8. Material development for ADS beam window

Three heats of 9Cr2WVTa steel have been smelted. The mechanical properties of the smelted 9Cr2WVTa steel have been investigated. It indicates that the C and Mn content as well as the heat treatment technologies affect the mechanical properties, therefore, the optimum element content and heat treatment technologies will be the key issues for the improvement of the 9Cr2WVTa steel. This research is being performed at moment. In order to get the martensitic structure and increasing its mechanical properties, the quenching treatment was performed. It can be seen that the black dots in the matrix increases when an increase in tempering temperature occurs, this may result from the increase of carbides with an increase of the temperature. There are little carbides in the matrix without tempering. The measurement results of the micro-hardness indicate that the hardness decreases with an increase of the tempering temperature, it may results from the dissolution of the martensitic under the increasing of the temperature.

Under the help of SCK and Brasimone, a few pieces of 9Cr2WVTa, 316LN and 12CrWTi have been tested in liquid Pb-Bi or liquid lead.

9. ADS related material compatibility study

The compatibility study is focused on the compatibility tests for the tungsten with water and sodium[14]. The compatibility tests for the forging tungsten with coolants have been performed in sodium at 500, 600 and 700 ℃, and in water at 100 ℃. The results show that a compact W_xO_y film was formed at the surface of the tungsten, its thickness is about 1–2 μ m. There are also corrosion product Na_2WO_4 on the surface of the specimens, the amount of Na_2WO_4 dependents on the temperature and the oxygen content in the sodium. After test for more than 400 h, the matrix of tungsten has not been attacked further by sodium and oxygen resulting from the protection of the W_xO_y film, the thickness of W_xO_y film and the weight loss become a constant. For the corrosion of the tungsten in water, a W_xO_y film at the specimen surface was formed at the beginning of the test, its thickness is about 0.8 μ m. This film is porous and loose, and they peeled off after test for more than 100 h. The new oxide film was not formed again because of the lack of the oxygen, the weight loss of the specimens is near a constant.

10. ADS related material radiation effects study

The spallation neutron source system is one of the three key parts of ADS, which provides source neutrons of about 10^{18} n/s for the burning of fuels. It is composed of the target and beam window. Stainless steels and tungsten are important candidate materials of the beam window and spallation neutron source target. They are irradiated by high-energy and intense protons and neutrons during operation. The accumulated dose could reach a couple of hundred dpa per year, and radiation damage is very severe in them. The radiation damage study of the spallation target and beam window materials is of great importance for the understanding of the lifetimes and safe operation of the ADS.

Dependence of radiation damage in the modified 316L stainless steel has been investigated on irradiation temperature from room temperature to 802 ^{o}C at 21 and 33 dpa and on irradiation dose up to 100 dpa at room temperature by the heavy ion irradiation simulation and positron annihilation lifetime techniques[15]. A radiation swelling peak was observed at about 580 ^{o}C where the vacancy cluster contains 14 and 19 vacancies and has an average diameter of 0.68 nm and 0.82 nm, respectively for the 21 and 33 dpa irradiations. It can be seen that before the peak temperature, the variation of positron lifetime τ_2 with irradiation dose increases with the increase of irradiation temperature. The higher the irradiation temperature, the larger the increase of lifetime τ_2 with irradiation dose. This indicates that the radiation damage depends on irradiation temperature more sensitively than on irradiation dose.

Before this experiment, radiation damage and its detailed thermal annealing behavior in α-Al_2O_3 irradiated at the equivalent dose, respectively, by 5.28×10^{16} cm^{-2} 85 MeV ^{19}F ions and by 3×10^{20} cm^{-2} $E_n \geq 1$ MeV neutrons have been investigated by the positron annihilation lifetime technique[16]. The experimental results show that all the positron annihilation parameters of lifetime and intensity in the heavy ion irradiated α-Al_2O_3 are in good agreement with the ones in the neutron irradiated α-Al_2O_3, and verify that heavy ion irradiation can well simulate neutron (proton) irradiation.

11. Conclusion

For long term and sustainable nuclear energy development, the ADS is an option in fuel circulation and energy generation. The ADS development has been started with a rather moderate project in China and it is still in the early stage. The goal for our ADS research is to establish the scientific and technological foundation for the future development of the ADS research step by step.

Acknowledgements The ADS research is supported by the China Ministry of Science and Technology (MOST) (Grant No. TG1999022600 and 2007CB209900)

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