High power particle accelerator for driving the nuclear waste transmutation system at nuclear power plant

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Abstract. The existing technology for treatment of NPP nuclear waste is a deep geological disposal technology. But this technology is still has possibility of leakage due to heating caused by radioactivity process from long lived elements of nuclear waste. The development of high power particle accelerator technology which can produce high power proton beam to induce nuclear spallation reaction has introduced a new accelerator-based technology for treatment of nuclear waste from NPP named the Accelerator-driven Transmutation Waste (ATW). This paper describes result of study on status of accelerator based transmutation system for NPP nuclear waste. ATW is subcritical nuclear reactor combined with high power proton accelerator for transmutation of nuclear waste utilizing neutrons from spallation reaction due to collision of high power proton beam with heavy metal material as spallation target. The types of accelerators used for ATW are cyclotron, linac and synchrotron which able to produce proton beam of 10 up to 100 MW beam power, 10 up to 100 mA beam current and 0.6 up to 1.5 GeV beam energy. Demonstration scale ATW requires 1 up to 2 MW of proton beam power to generate 50 up to 100 MWth of thermal power in subcritical reactor. Whereas industrial ATW requires 10 up to 75 MW of proton beam power to generate hundred MWth of thermal power in subcritical reactor. Several countries have roadmap of ADS research and development which the main goal of nuclear waste transmutation using ADS called ATW. ATW also can be used for electrical energy production which is safer and more reliable.

1. Introduction
Nuclear power plant (NPP) is the implementation of nuclear technology for energy generation. It is undeniable that beside electrical energy an NPP also produces nuclear waste which contains transuranic nuclides and radioisotopes of long-life fission products with a half-life of $10^5 - 10^6$ years and high radiotoxicity. The radiotoxicity of nuclear waste can be reduced by the transmutation method which is carried out by using high flux neutrons in a nuclear reactor. The high flux neutron irradiation induces the fission reactions in nuclear waste so that the radionuclides contained in waste decay into stable nuclides. In this case, the fuel of the nuclear reactor used for transmutation must be free from uranium to prevent the reformation of long-life nuclides. Therefore the nuclear reactor must be operated in a subcritical mode[1].

A subcritical reactor that is used for the transmutation of nuclear waste needs a neutron source which can produce a very high flux of neutron. Such neutron source can be realized by a spallation reaction that occurs when energetic particles e.g. protons, deuterons, neutrons, etc. hit a target made of a heavy metal element. The spallation reaction needs a proton beam with energy of several hundred MeV which can be produced by a high energy particle accelerator. If the proton energy is above around 1 GeV, then...
the neutron yield per incident proton is proportional to the proton energy. In the proton energy range of 1 - 2 GeV there are around 20 - 30 neutrons produced by each incident proton[2].

The combination of a subcritical reactor and a high energy particle accelerator is known as Accelerator Driven System (ADS)[3]. The facility for the transmutation of nuclear waste which uses ADS technology is known as Accelerator-driven Transmutation Waste (ATW)[4]. In ATW, the partition process of NPP spent fuel is done, and then followed by the transmutation process in the subcritical reactor using the neutrons from the spallation reaction (known as spallation neutrons). The ATW is also known as a hybrid reactor because besides the ATW is used for transmutation, it can also be used for electric production[5]. This paper presents the result of the study on the principle of a nuclear waste transmutation system based on the accelerator technology (ATW) and some requirements which must be fulfilled either by the particle accelerator or by the subcritical reactor assembly. Several examples of ongoing ADS project in the world related to the nuclear waste transmutation are also presented in this paper.

2. Methodology
The research was carried out by the literature study method. It aims to study the nuclear waste transmutation system that uses the combination of the particle accelerator technology and subcritical reactor known as Accelerator Driven System (ADS). Therefore the literature study covers the materials related with the application of ADS for the transmutation of NPP nuclear waste which includes the NPP radioactive waste, the NPP spent fuel, nuclear waste transmutation, the working principle of ATW, proton accelerator for ATW, spallation target, subcritical reactor, and classification of ATW. The materials for the literature study were obtained from journals, proceedings, articles, and other relevant information which can be accessed via the internet.

3. Result and Discussion
3.1. The transmutation of nuclear waste using particle accelerator
Transmutation is one of the methods to decrease the radioactivity level of nuclear waste, where nuclear waste consists of long-life nuclides is transmuted into a more stable or short-life nuclide. Transmutation can be defined as the transformation of a material into a new material by changing the structure of the nuclei, so it is often called as a nuclear transmutation. Nuclear transmutation was known for the first time when Rutherford in 1919 used natural α particle to convert nitrogen element $^{14}$N into oxygen $^{17}$O through the reaction[6]:

$$^{14}_7N + \alpha \rightarrow ^{16}_8O + p$$  (1)

With the advent of particle acceleration using an electric field, Cockcroft and Walton in 1932 carried out the transmutation of lithium $^7$Li into helium $^4$He using accelerated proton with the energy of ~ 800 keV. This is the first artificial transmutation which the nuclear reaction can be written as[6]:

$$^7_3Li + ^1_1H \rightarrow ^4_2He + ^4_2He + 17,2 MeV$$ (2)

The progress of charged particle accelerator using the technique of DC high-voltage, RF circular (cyclotron), and RF linear (linac) has brought to the capability to accelerate electron, proton, and other charged particles until their energy exceeds the nuclear binding energy of target. These energetic particles induce nuclear reactions that create new elements that are very useful for the transmutation and fission processes. In a nuclear reaction in which the energy of the colliding particles exceeds the nuclear binding energy of the target nuclei, it is always accompanied by the emission of neutrons and other particles. Thus, the particle coming from accelerator serves as secondary neutrons generator which can be used in various ways. One of them is for the transmutation of the isotope of natural uranium U-238 and natural thorium Th-232 into the fissile plutonium Pu-239 by the following reactions[6]:
These fissile isotopes are separated by the process of irradiated fuel and refabricated as a new fuel containing the Pu-239 and U-233 to be used in a power nuclear reactor. This process is known as the accelerator breeder system, but it has not been implemented in commercial-scale because the cost of the produced fissile material is very expensive compared to the cost of the enriched uranium. The economical cost of the nuclear fuel produced by the accelerator breeder depends on the cost of the neutron production for the above transmutation reactions and the cost of the separation process of fissile isotopes from the origin species[6].

3.2. The working principle of ATW system

A subcritical nuclear reactor facility for the transmutation of nuclear waste by using a high power particle accelerator as its neutron source is known as Accelerator-driven Transmutation of Waste (ATW). This technology arises to facilitate the transmutation process of spent nuclear fuel. The main components of the ATW system is schematically illustrated In Figure 1[7].

They consist of: a high power proton accelerator (linac or cyclotron) for producing proton beam with energy of 1 - 1.5 GeV or beam power of several MW, a facility for separation process of the spent fuel before it is baited as fuel in the subcritical reactor assembly and a subcritical reactor as transmutation device which is known as transmutter or blanket, in it there is a spallation target as a neutron source.

The ATW uses neutrons produced by the spallation reaction between a high energy proton beams with a target or blanket consists of fissionable fuel and radioactive waste materials. The blanket is like a reactor in which the fission process constitutes the power supply but does not like the conventional reactor. The blanket constitutes a subcritical reactor so that the chain reaction cannot be sustained without an external neutron source. The spallation neutron source constitutes an external neutron source in the ADS system. ADS can use uranium, plutonium or thorium as the fuel.

The spent nuclear fuel in the ATW system is reprocessed in the separation facility using the pyro chemical process in which the uranium and most of the fission products are separated from the TRU elements and the long-life fission products. Further, the TRU elements and the long-life fission products are processed to be fuel and blanket in the subcritical reactor. The transmutation takes place in the

\[
{^{238}}_{92}U + \frac{1}{0}n \rightarrow {^{239}}_{92}U \rightarrow {^{239}}_{93}NP + \beta^- \quad \text{and} \quad {^{239}}_{93}NP \rightarrow {^{239}}_{94}Pu + \beta^-
\]  

\[
{^{232}}_{90}Th + \frac{1}{0}n \rightarrow {^{233}}_{90}Th \rightarrow {^{233}}_{91}Pa + \beta^- \quad \text{and} \quad {^{233}}_{91}Pa \rightarrow {^{233}}_{92}U + \beta^-
\]
subcritical reactor triggered by an accelerator in which the TRU elements and the long-life fission products are placed around the spallation target.

The spallation neutrons are multiplied by the fission process around the target so that the neutron flux becomes high and fulfils the transmutation requirement. The transmutation aims to destroy the TRU elements and the long-life fission products. The ATW system also produces heat that can be used for the production of electrical energy, so it is also known as hybrid reactor. A little bit (10 - 20%) of the electrical energy is used to operate the accelerator, while the rest is distributed to the grid for the other need of electrical energy\cite{7}\cite{8}.

3.3. Proton accelerator for the ATW system

The proton accelerator in an ATW system is used to produce a proton beam that will bombard the target to induce a spallation reaction and subsequently produce neutrons. The proton accelerator used for the ATW system has to fulfil the following requirements\cite{9}:

a. The proton accelerator should be a high power accelerator with the beam power of several MW.

b. The proton accelerator should be a high energy accelerator with the beam energy of 600 MeV up to 1.5 GeV.

c. The proton accelerator should have a very high proton beam stability which includes the beam energy stability of ± 1%, the beam intensity stability of ± 2%, and the beam size stability of ± 10%.

d. The proton accelerator should have a high reliability in which the number of beam trips is less than 5 times per year.

e. The proton accelerator should have a very low beam loss, i.e. not more than 1 Watt/m.

Based on the above requirements there are several types of proton accelerator which can be utilized for the transmutation of nuclear waste due to the capability to provide a proton beam used to trigger the transmutter, namely:

a) Cyclotron: an oldest type of accelerator and mostly has been used for the low beam energy and beam power applications. Even so, a cyclotron has experienced evolution so that it can achieve high beam energy and beam power. For example, the cyclotron at the Paul Scherer Institute (PSI) in Switzerland has achieved a continuous proton beam power of 1.3 MW at the energy of 0.59 MeV, and the cyclotron has been upgraded so that it can achieve the proton beam power of 2.4 MW\cite{10}.

b) Linear accelerator (linac): a type of accelerator in which the trajectory of the particle is straight or linear. A linear accelerator consists of several sections depend on the required particle energy. In each section a particle passes only once towards the next section. Linac is applied in many fields because it has several excellences, i.e. it is easy to inject and to extract a beam due to an open geometry, easy to transport a high flux beam, and it has a high duty cycle. There are 2 types of linac which can be used for transmutation of nuclear waste, namely\cite{9}:

- Normal conducting proton linac, for example the LANSCE linac at the Los Alamos National Laboratory USA which can produce pulse proton beam with a proton beam power of 1 MW,
- Superconducting proton linac, for example the SNS linac at the Oak Ridge National Laboratory USA which can produce pulse proton beam with a proton beam power of 1.1 MW.
c) Synchrotron: a type of accelerator which is the further development of a cyclotron. Synchrotron uses a time-variable magnetic field as a deflecting device which is synchronized with the particle energy so that the radius of the particle is relatively constant although the particle energy is increasingly high. A synchrotron can produce a proton beam with a beam power of more than 1 MW but is limited to pulse operation at relatively low duty factor[9].

The choice of accelerator type depends on the need of the proton beam power for driving the subcritical assembly. The high power requirement of the ATW leads to a preference for a linac than a cyclotron. The reason is that linac has capability to accelerate protons up to several GeV with an intensity of more than 100 A. It indicates that linac can produce a continuous proton beam in the range of a hundred MW. While the maximum energy of proton beam produced by a cyclotron is limited of ~ 1 GeV with a maximum intensity of ~ 10 mA due to the relativistic effect[11].

3.4. The spallation target
The spallation target constitutes a primary neutron source of ATW. The neutrons are produced by the spallation reaction in which a light projectile (proton, neutron or light nuclei) with a kinetic energy of several hundred MeV up to several GeV interacts with heavy nuclei (such as Pb), and causes the emission of large numbers of hadron which mostly are neutrons. The number of spallation neutrons formed by each proton that hit the spallation target depends on the proton energy and the target material[12].

The spallation target is designed in such a way to generate the spallation neutrons as many as possible, as well as to evacuate the heat generated by the spallation process. The thermal load of the spallation target is very large due to the conversion of 10 MW protons into neutrons. Therefore it should be cooled, generally by using gas, heavy water or liquid metal. The damage of window and other components caused by the proton radiation also should be considered. In the proton energy range of several GeV, the damage caused by the proton radiation is not significantly affected by the proton energy. It is favourable by decreasing the beam current and increasing the beam energy. The best choice of material for the spallation target according to the requirements is molten lead or lead-bismuth eutectic (LBE). But the significant problem of LBE is the production of radioactive polonium and the highly mobile high energy proton reaction with a neutron in bismuth. At high temperature, the polonium contained in LBE will be released quickly. While Pb produces a few of polonium, but its operating temperature is higher.

The spallation target can be made of solid or liquid material. The solid target consists of solid material in the form of rods, spheres, or plates to produce the neutrons, and coolant flowing between the elements for heat removal. Tungsten, tantalum, and lead are the solid materials that are mainly used for solid spallation target. Neutron yield (n/p) is proportional to the atom weight of the target and the proton energy (linear for the proton energy range from 0.8 up to 2 GeV). The liquid target uses a flowing liquid metal that acts both as the source of neutrons and the heat removal media. The liquid mercury and lead-bismuth eutectic (LBE) are the liquid materials used for liquid spallation target[9].

3.5. The Subcritical Reactor
Subcritical reactor is the main part of the ATW system in which the nuclear waste transmutation process takes place so that it is frequently known as transmutter. The transmutation takes place due to the neutron bombardment on nuclear waste material. Neutrons are produced by the spallation target which then amplified by the fission process. Besides that, a neutron absorption process causes a decrease of the neutron population. In a critical reactor, there is a balance of neutron increase and neutron decrease so that a steady-state condition is achieved (the reactor criticality value $k_{eff} = 1$). In a subcritical reactor, a steady-state condition also can be found provided that there is an external neutron source. In this case, the neutron population from the external neutron source is amplified by a factor $[1/(1 - k_{eff})]$, where $k_{eff} < 1$ is the level of subcriticality. The total population of neutron or the level of reactor power is controlled by the intensity of an external neutron source. In this case, the spallation target constitutes the external
neutron source. At a constant subcritical level, the reactor power is determined only by the proton beam intensity.[2]

The spallation target is installed in the centre of the subcritical reactor so that the produced spallation neutrons are amplified by the subcritical reactor through the fission process. Quantitatively, the total fission in the subcritical reactor \( N_{fiss} \) can be expressed as follow[3]:

\[
N_{fiss} = N_h \Gamma_h \frac{k_{eff}}{(1 - k_{eff}) \nu}
\]

where \( N_h \) is the number of fission due to the reaction with a high energy proton, \( \Gamma_h \) is the number of neutrons produced per fission due to the reaction with a high energy proton in the subcritical blanket (spallation, evaporation, and high energy fission), \( \nu \) is the number of neutron per regular fission, and \( k_{eff} \) is the amplification factor of neutron produced by regular fission.

The proton beam current that plays a role in maintaining the criticality of a subcritical reactor can be reduced by increasing the \( k_{eff} \) value. If the \( k_{eff} = 1 \), so the blanket becomes critical and it needs no more the external neutrons (spallation neutrons) generated by the proton beam. From the results of several studies it is concluded that for the incineration process of the minor actinide, the suitable value of \( k_{eff} \) is from 0.9 up to 0.985. It should be noted that the \( k_{eff} \) value depends only on the characteristics of the reactor core (i.e. dimension, shape, material composition, temperature) and depends not on the characteristics of the source. The material composition consists of the mixture of nuclear fuel, cooling material, structure material, and controller material. For the sustainability of the chain fission reaction, the materials should be constructed such that the configuration is suitable with the sufficient dimension and the correct shape[13].

A subcritical reactor usually uses uranium fuel or thorium fuel. The advantage of using the thorium fuel in the subcritical reactor is that it produces the long-life actinide radioactive waste less than when using the uranium fuel. But thorium is not a fissile material so it should be converted into \(^{233}\text{U}\) by the neutron irradiation. The thorium fuelled ATW can be designed such that the long-term change of the reactivity caused by the burn-up is minimum. By using the variable proton current the need for parasitic absorber for the long-term reactivity control can be omitted. The ATW system is expected to have the breeding characteristic which is superior compared to the critical reactor[13].

Accelerator driven subcritical reactor has two principal advantages relative to critical reactor. First, it has greater flexibility with respect to fuel composition, and second, its safety is potentially could be enhanced. Accelerator driven subcritical reactor is suitable for burning fuels that would degrade neutronic characteristics of the critical core to unacceptable levels due to small delayed neutron fractions and short neutron lifetimes. From the standpoint of critical reactor operation, this condition would be problematic. It should also be noted that the reactor power maintenance could be simplified because the reactor power is proportional to the accelerator current[14].

3.6. The Classification of ATW System

The ATW system can be classified into 2 groups, namely, the demo scale and the industrial scale ATW systems[9].

a) The demo scale ATW system

The demo scale ATW system is a facility to demonstrate the transmutation process in a flexible research facility in which the subcritical core is coupled with a high power proton accelerator of MW power. The demo scale transmutation of nuclear waste needs a beam power of 1 - 2 MW to produce a thermal power of 50 – 100 MWt in a subcritical assembly, depends on the neutron amplification factor of the subcritical assembly. It can be realized by using a continuous (CW) or pulse proton beam in the beam energy range from 0.5 up to 3 GeV which can be implemented using a cyclotron or superconducting linac. The example of the demo scale ATW system is the MYRRHA (Multipurpose hYbrid Research Reactor for High-tech Applications) in Belgium which aims to research and development of nuclear waste
transmutation. The facility uses the 600 MeV proton accelerator which can deliver a beam power of 1.5 MW to the subcritical assembly with thermal power of 85 MWt.

b) The industrial ATW system
The industrial ATW system is a facility of nuclear waste transmutation which has been costly and technologically optimized. The industrial-scale transmutation of nuclear waste needs a beam power of 10 up to 75 MW to produce thermal power of the order of GWt in a subcritical assembly. It requires a continuous proton beam with an energy range 1 – 2 GeV which can be provided only by the superconducting linac. In that energy range, the proton beam can produce spallation neutrons with an optimum neutron yield. The examples of industrial-scale ATW systems are (a) the JAEA transmutation facility: beam energy of 1.5 GeV, beam power of 30 MW, and thermal power of 800 MWt, (b) the European Facility for Industrial Transmutation (EFIT): beam energy of 0.8 GeV, beam power of 16 MW, and thermal power of several hundred MWt, (c) Los Alamos Accelerator Transmutation of Waste (LA-ATW): beam energy of 1 GeV, beam power of 45 MW, and thermal power of 840 MWt.

3.7. The Status of ADS (ATW) Research and Development
Since the late 1980s, the research and development of ADS for the transmutation of the long-life radiotoxic nuclei and the production of nuclear energy is more and more increase throughout the world. The research and development of ADS in several countries are conducted with a very different policy of the nuclear fuel cycle. France, Japan, and Russian have the policy of the nuclear fuel cycle to reprocess plutonium (recirculation). USA and Sweden have the policy of one through nuclear fuel cycle. Even in countries like Italy and Poland which have not any NPP the research of ADS is still conducted. The great interest in the research of ADS for transmutation like in France and Japan is triggered by the new problem related to the future fast breeder reactor[7].

USA, Europe Union, Japan, and other countries have designed the roadmap of ADS research and development, and have researched the appropriate nuclear waste. The status of the ADS/ATW research and development in some countries are summarized below:

3.7.1. In 1999 the US Department of Energy developed a roadmap for ATW program. The Advanced Accelerator Applications which has been already started in 2001 aimed at the development of technology for the transmutation of transuranic (TRU) and long-lived fission products, and to build an accelerator-driven test device in 2010. The ADS related activities in the US are conducted at the Los Alamos National Laboratory (LANL), such as the development of a high power (several hundred MW) LINAC under the Accelerator Production of Tritium (APT) program. The program aimed to reduce the number and risk of long-lived spent nuclear fuel from the reactors in the US. Now the ADS study has been an integral part of the US advanced nuclear fuel cycle system[15].

b) The European Union regards ADS as one of the key technologies for nuclear waste management. The Technical Working Group led by Carlos Rubbia, the scientist from CERN, to develop a research and development program framework. The framework presents the EURopean Programme for the TRANSmutation of High Level Nuclear Waste in an Accelerator Driven System (EUROTRANS) program planned to form the design concept of the eXperimental Transmutation in an Accelerator Driven System (XT-ADS) and the European Facility for Industrial Transmutation (EFIT). Belgium is one of the European Union countries that adopted the corresponding national research program from the program framework by proposing the Multi-purpose hYbrid Research Reactor for High-tech Applications (MYRRHA). MYRRHA is an accelerator-driven Pb-Bi cooled fast neutron subcritical reactor system scheduled to run in 2023 for the research of materials, nuclear fuels, radioisotopes production, nuclear waste transmutation, and fundamental research. A 600 MeV, 0.1 - 4.0 mA proton superconductor LINAC serves to produce high power proton beam (up to 2.4 MW) with an extreme reliability level (MTBF) of more than 250 hours[16][17].
c) In Japan the investigation of ADS for transmutation of minor actinide (MA) which will be partitioned from the high level waste (HLW) is conducted by the Japan Atomic Energy Agency (JAEA). The Option Making Extra Gain from Actinides (OMEGA) program was launched for the final disposal of nuclear waste. The long-term program of the partition and transmutation of nuclear waste, including the study of the physical and chemical characteristics of MA and long-lived fission products (LLFP) has been conducted. The technical specification of investigated ADS is a subcritical reactor with 800 MW thermal power cooled using lead-bismuth eutectic (LBE). A 1.5 GeV, 20 mA proton LINAC is applied to generate spallation neutrons in the subcritical reactor. This ADS can transmute about 250 kg MAs per year[18].

d) India also has interest in developing ADS for transmutation of nuclear waste. The effort to develop 1 GeV high intensity CW proton accelerator has been initiated by conception of accelerator development program which is conducted in 3 phases[19]: (a) Phase 1: a 20 MeV, 10 mA normal conducting front-end called the Low Energy High Intensity Proton Accelerator (LEHIPA). It is now under commissioning, consists of a 50 keV proton ion source, 3 MeV room temperature Radio Frequency Quadrupole (RFQ) followed by Drift Tube LINAC (DTL) to 20 MeV, (b) Phase 2: a 200 MeV, 10 mA, superconducting accelerator using single-spoke resonators, called the Medium Energy High Intensity Proton Accelerator (MEHIPA), (c) Phase 3: The full 1 GeV, 10 mA, CW, High Energy High Intensity Proton Accelerator (HEHIPA) that will use elliptic cavities from 200 MeV to 1 GeV.

e) Since the 1990s, China has developed research on ADS concept. The China Institute of Atomic Energy has built the fast-thermal coupling ADS subcritical platform named Venus No.1. In 2011 the Chinese Academy of Science (CAS) started the ADS strategic research priority program aimed at verifying the ADS principle and solving the key technical issues in the accelerator, spallation target, and reactor system. For the future development of nuclear energy requirement, China proposed the concept of Accelerator-driven Advanced Nuclear Energy System (ADANES)[20].

4. Conclusion
The progress in the technology of high power particle accelerator gives a chance to apply proton accelerator for solving the problem of NPPs nuclear waste using Accelerator-driven Transmutation of Waste (ATW) system. The accelerators which can be used for the ATW system are cyclotron, linac, and synchrotron. They can produce a proton beam with a beam power of 10 up to 100 MW, beam current of 10 up to 100 mA, and beam energy of 0.6 up to 1.5 GeV. The demo scale ATW needs a proton beam power of 1 up to 2 MW to generate a thermal power of 50 up to 100 MW_{th} in the subcritical reactor. While the industrial scale ATW needs a proton beam power of 10 up to 75 MW to generate a thermal power of several hundred MW_{th} in the subcritical reactor. The transmutation using the ATW system is safer than using the critical reactor system because the control system and power alteration are triggered by an external neutron source i.e. spallation neutron source. The research and development of ADS in several countries are conducted with a very different policy of the nuclear fuel cycle. They have designed the roadmap of ADS research and development, and have researched the appropriate nuclear waste.

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