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MAIN CRITERIA AND REQUIREMENTS TO THE PROCESS OF NUCLEAR TRANSMUTATION OF HIGH LEVEL WASTES AND TO THE INSTALLATIONS FOR NUCLEAR TRANSMUTATION

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I. INTRODUCTION.

Nuclear transmutation (NT) is an alternative approach to the problem of high level wastes (HLW) management in atomic industry. NT is based on a number of physical and chemical processes involving possibilities of the prospective nuclear technologies of the 21-st century, realization of these possibilities would resulting in the following:

1. Reduction of general and specifie HLW activities and times for HLW controlled storage.
2. Utilization of some useful radionuclides in isotops technologies as well as in thermal and electrical energy production.
3. Enhancement of atomic energy safety.
4. Use of uranium 238 for production of thermal and electrical energy.

All these attractive possibilities could be realized as a result of HLW transformation (into short lived or stable nuclides) after interaction with a beam of neutral or charged particles. NT of HLW is characterized by multi stage transformation of nuclides and their multi branching in particles interactions with radioactive nuclei. Even in case of fission reaction ($n_t f$) we can't speak about one stage process of incineration because after fission the nucl ide gives off several fragments (fission products) which undergo subsequent transformation (transmutation). Analyses of physical processes of NT and nuclear physical properties indicate the following possibilities of HLWNT:

- to make use of nuclear reaction (n,γ) and of the subsequent neutrons multi capturing (xn,γ) resulting in stable nuclei formation or in formation of radioactive nuclei subject to electron or positron decay, isometric transition or electron capture.

- to use threshold reactions (n,p) and (n,α) to separate chemically parent and daughter nuclei.
- to use nuclear reactions $(n, 2n)$ and (n, xn) for high energy neutrons.
- to use reactions (γ,n) possessing as a rule energy thresholds.
- to use nuclear reactions of fission (n,f).

The pointed out reactions can be realized in nuclear energy plants with intensive neutron fluxes. -To such plants we refer thermal and fast reactors and charged particles accelerators (proton, deutron, triton, etc.) with appropriate convertors. Reactions (γ,n) can be realized in electron accelerators. Finally there is a very attractive possibility to use for NT of HLW charged particles beam.

However, if a nuclear reactor is used as a NT installation then in the reactor core housing nuclides for transmutation the above mentioned reactions (n,γ), (n,p), (n,α), $(n,2n)$, (γ,n) and (n,f) can be realized. Firstly, it means that there is a probability to realize one or several nuclear reactions depending on the reactor core conditions and nuclear cross sections. Secondly, nuclear properties of parent and daugther nuclides can be different and it would being about different types of nuclears transformations. In addition a daughter radionuclide can undergo a certain radioactive decay and the decay products in their turn can interact with the neutron flux. It is clear that NT process has at least 3 stages: capturing (stage 1); $\beta^{\pm}$ - daughter nucleus decay (stage II), provided that neutron capturing by the daughter nucleus can be neglected due to very small interaction cross-sections; neutron capturing by the products of the daughter nucleus disintegration (stage 111). However this process can have 4 stages if the daughter products have large interaction cross-sections and along with radioactive decay there occurs intensive neutron capturing. Schematically these processes of mutual transformations are shown in Fig.1, where Xi is parent nucleus; Yi - daughter nucleus; Z_i - nucleus produced after decay of nucleus Yi; (n,γ) - indicated above nuclear reactions with neutrons. Fig.1 shows that the number of transformations can be enough large. This scheme will become more complicated if there are several isotops of the parent radionuclei Xi , X_2, X_3 . . . . In Fig.1 we can also see that during NT process there can be multi branching. In this case there is a number of quite complex optimization problems, e.g. for minimum values of Z_1 and Z_2 or for minimum values of the absorbed neutrons, etc. .

2. POSSIBLE DIRECTIONS IN CREATING INSTALLATIONS FOR NT of HLW.

There are 3 possibilities to create powerful neutron sources: fission, nuclear reactors, charged particles accelerators and thermonuclear reactors. What factors are to be taken into consideration if we wish to define priority type of the NT installation to be developed? We believe that the following criteria should be considered (some of them are quite obvious nevertheless let us mention them as well]:

1. NT energy efficiency
2. NT rate
3. NT process safety
4. Maximum use of time tested technologies and designs
5. Cost efficiency.

Comparative analyses of the above mentioned types of NT installations must be done at a later stage of the concept study but already today there is enough data to chose priority directions for further technical developments. The researches carried out at. ITEP have shown that at the present level of technologies a complex for NT of HLW containing a proton accelerator, target-convertor and heavy water blanket seems to be the most promising. However when taking decision on the type of NT installation to be prefered and technical equipment for such installations to be developed some other criteria are to be taken into consideration as well and it seems appropriate to analyze these criteria here at some length.

3. GENERAL CRITERIA.

It is quite obvious that recommending NT process as an alternative approach to the HLW management we must firstly answer the question natural for any expert: what are the advantages of NT of HLW when compared with HLW controlled storage or HLW deep geologic deposition.

To answer this question we must make comparative analyses of technical and economical features of different approaches the HLW management. Although the jobs or the analyses are not quite completed in our country yet it is possible now to indicate some essential criteria to be taken into consideration when making such comparative analyses.

1. NT technological processes and installations for NT of HLW must not affect the environment. HLW NT effective process must contribute to reduction of the total activities of the HLW and consequently lead to reduction of collective radiation doses and risks of human affection.

Two aspects are meant here: normal situation and an emergency and their technical and radiation effect on the environment. Possible radiation affection can be illustrated by the following example. Let us take 2 illustrating for transmutation: a proton accelerator complex with a subcritical blanket and a fast reactor. Capacity of actinides loading into the blanket is 10 ÷ 20 times less than that in the fast reactors. It means that radiation hazards from the accelerator complex are at least one order of magnitude less then those from the fast breeder. But to estimate population collective radiation doses is not that simple due to the each of reliable data on different ways of HLW management. That is why our estimations are very relative. Nevertheless calculations show that there is a possibility to reduce HLW activity to the level of uranium natural activity. That is why we can say with a certain degree of confidence that possible radiation hazards from HLW will be considerably reduced. As for technological affection we must bear in mind that there is a possible energy in the process of NT of HLW. The last statement suggests the second criterion.

NT process must not worsen the balance of energy generated by nuclear fission uranium or plutonium in NPP reactors,

It is quite natural that many researchers are investigating NT possibilities in reactors power plants with thermal and fast reactors at IAERI (Japan) they have considered NT in PWR,HTJR and BR reactors. IPE (Moscow) have made calculations for RBMK reactors as well as for modified reactors with lead coolant. FEI (Obninsk) is studing possibilities of actinides transmutation in fast reactor BN-800 and a special fast reactor with sodium coolant. At ITEP we have considered a special heavy water reactor with thermal capacity of 1000 Mwt Cit was reported about this reactor at the Obninsk International seminar 17.1-5.19911 as well as a modular heavy water reactor MTR-500 designed for thermal energy production. The calculations show that for HLW NT in reactors with thermal neutrons a larger amount of enriched uranium is required. At the same time it was shown that reactor MTR-500 due its design features allows technetium 99 transmutation without affecting the nucler fuel burning up.

However a proton acceleration complex with a subcritical blanket and fast burner reactors have positive energy balance. It means that they have advantages when compared to thermal neutrons power plants reactors.

4. Installations for NT of HLW must be of enhanced safety. Any risk of an accident caused by supercritality with instant neutrons flux increase must be completely ruled out. Inherent safety features and passive safety systems must be made full use of and all necessary steps to exclude radioactivity spread outside the installation building must be taken. No explosive equipment and technologies must be used.

This general requirement is eventual because in the irradiated volume of the installation for NT there is a considerable of long-lived, fission products and actinides exceeding much this amount in NPP reactors. From this point of view an acceleration complex with a subcritical blanket with $K_{eff} \leq 0.95$ seems to be more preferable. The available today data on the safety properties of fast reactors with sodium coolant show that they do not satisfy this criterion. According to the calculations for the fast burner reactor independently made by FEI (Moscow) and JAERI and with CRIEPI specialists (Japan] sodium void reactivity coefficient is positive and makes about 4 ÷ 4.5 %.This is a serious drawback of fast reactors with sodium coolant, From this point of view the fast reactor with lead coolant being developed by IPE can offer higher radiation safety. It. must be mentioned that IAEA regulations do *not* prohibit exploitation of NPP reactors with positive feed backs but our regulations after Chernobyl accident rule out exploitation of such reactors. For safety reasons NT installations can be made underground if necessary,

4, Installations for NT of HLW must be integrated into a closed nuclear cycle. If HLW of the military atomic industry are to be eliminated the technological cycle of wastes transmutation must be closed and contain a radiochemical plant for reprocessing of the irradiated target materials as well as for fission products and actinides fragmentation. In any nuclear cycle special storages for fragmentated fission products and actinides must be provided for several decades (30÷40 years].

The requirement for the nuclear fuel cycle to be closed is necessary for realization of HLW NT processes. It is quite obvious that without radiochemical reprocessing NT realization is impossible. In the military atomic industry HLW are produced as a result of fuel elements reprocessing and are as a result stored in a liquid form without separation. And this necessitates development of effective technologies for their separation. The last requirement as well as the necessity to have minimum radioactivity accounted for by admixtures in the HLW under transmutation helps us to formulate the next criterion.

5. Purification coefficients in the process of HLW partitioning must not be less than 99.9% and losses during radiochemical reprocessing of the irradiated target materials must not be more then 0.1% of their weight.

To realize values indicated in this criterion is quite problem. However we must keep in mind that the lower is the amount of accompanying admixtures the lower is the radioactivity accounted for by them. Experiments made at Chlopin Radium Institute demonstrate that there are technical possibilities to satisfy this criterion.

6. It is desirable to place installations for NT of HLW in the immediate proximity from radiochemical plants for reprocessing of the irradiated target materials with HLW to be transmutated and preparing the HLW for transmutation in the form required,

This requirement is necessary due to 2 reasons. First we exclude HLW (especially liquid one) transmutation and will have no transport accident. Second it is always very difficult to get new industrial areas especially for industrial sites with considerable concentration of HLW with high total activity due to the public objections.

7. After NT the remnants of wastes with specific radioactivity not higher than that of the natural uranium must be hermetized.

HLW NT process envisages HLW irradiation in the installation irradiation volume, on site radiochemical separation of the isotops produced, radiochemical reprocessing of the target materials after irradiation completion. At all stages of radiochemical reprocessing there will be losses of fission products and actinides and it must be taken into consideration when defining the total radioactivity balance. This makes the problem of fission products and actinides burning up. It is necessary to define the number of cycles for the target materials irradiation and number of reprocessing and to determine the total amount of losses during these technological operations as well as the volume of the remaining radioactive wastes to be deposited.

8. The process of HLW NT must be economically profitable.

The requirement may seem to contradict the common sence at the first glance as it is well known that incineration industrial wastes always requires considerable financing. The point, is to consider the process of HLW NT not only as incineration of HLW with the help of an intensive neutron source but as a multipurpose process.NT can be presented as a complex of following technological processes:

1. Partitioning of long-lived HLW and use of separated radionuclides in industries, farming, medicine and science thus stimulating further development of radionuclides technologies. We must keep in mind here the prospective demands for radionuclides in the 21-st century, e.g. for such as cesium, praseodium, technetium. In principle we can consider the possibility of returning zirconium-93 into the nucler fuel cycle for fuel elements manufacturing.

2. HLW transmutation (after partitioning) into useful radionuclides For example transmutation of neptunium-237 in plutonium-238 to be used for energy production. Such a possibility was considered in one of reports presented at the Obninsk International Seminar 07.1-5.91.

3. Production of thermal and electrical energy in the process of

NT of actinides.

4. Use of y-radiation energy for the processes of polimerisation, sterilisation, etc.

5. Use of intensive neutron sources for research in the field of material study and solid physics.

6. Use of uranium-238 which is present in large amounts in the NPP wastes after separation for production of new fuel elements.

Such multipurpose approach will allow to reduce capital investments and exploitation costs for HLW NT installations.

All said above allows us to formulate the final requirement.

9. Costs of installations for HLW NT must be minimal.

It is obvious that the costs will be defined by a number of factors such as:

- NT process energy efficiency
- high efficiency of HLW NT process
- nuclear technical and radiation safety of HLW NT process as well as of NT installations
- degree of purification (partitioning) of radionuclides
- costs of technological operations and equipment for HLW NT, etc.

The costs required must be defined at the further stages of development.

Besides the general criteria we can indicate a number of requirements for technical equipment. These requirements must be specified at a later stage but even now we can formulate them for the accelerator complex suggested by ITEP.

4. REQUIREMENTS to TECHNOLOGICAL EQUIPMENT of the ACCELERATOR COMPLEX.

As said above the accelerator complex for HLW NT consists of 3 main units: a high flux proton accelerator, target-convertor and heavy water blanket. Each unit has supporting system as well as systems of maintenance, control and diagnostics. In each unit the process of energy transformation is taking place. In the linear accelerator electromagnetic energy is transformed into kinetic energy of the moving protons. In the target the protons kinetic energy is transformed firstly into kinetic energy of the primary particles (neutrons, p-mesons, γ-quants) and spallation products; and secondly into thermal energy. In the blanket step by step transformations of kinetic energy of the primary irradiation into kinetic energy of the second order particles and fission fragments into thermal energy and then with the help of the thermomechanical system into electrical energy takes place. Physical processes in the units of the accelerator complex have their characteristic features and times which must be taken into consideration when designing such a complex, However besides differences there are close interconnections both direct and indirect among the indicated units of the complex and they must also be taken into consideration to ensure normal exploitation of the complex.

We can formulate the following requirements to exploitation of the complex and to its main units designs:

srtuctures and exploitation modes of the accelerator complex must be designed to ensure perfect matching of the main units. Means must be provided to allow possible dispatching location in each unit as well as in supporting systems and to rule out any possible negative effect of the units on each other.

Although this requirement is quite obvious it should be mentioned in the first place because to ensure tuning of 3 complicated units of the accelerator complex with a great number of supporting systems is a rather complicated undertaking. We can site the following examples of direct and indirect effect of the units on each other. Normal exploitation of the liner accelerator with the given rated parameters define totally the operation modes for the target and blanket. Any deviation in the accelerator parameters bring about immediately deviations in the operation modes of the target and blanket. At the same time during the accelerator complex operation heavy metal vapours and spallation product from the target will enter the vacuum chamber of the junction block, and certain measures must be taken to prevent the vapours entering the accelerator vacuum chamber and to affect the proton beam and the accelerator structures. We can take another example when deviation in the blanket mode of operation will necessitate immediate switching off of the accelerator. The list of such examples can be continued, To satisfy the requirement of matching the operation of the 3 main units we must develop systems and algorithms of control for conventional and emergency situations.

However besides matching of the operations of all units and systems of the accelerator complex its actual parameters must correspond to those of the design project for transmutation of the necessary radionuclide or radionuclides group. The concept of the accelerator complex developed at ITEP envisages transmutation of certain fission products and actinides and the problem arises to optimize position and irradiation conditions of the target materials in the blanket at the given proton beam parameters. All above can be summed up as the following requirement

- Actual parameter-s of the accelerator complex, its units and systems must correspond to the design project parameters optimized for the preselected transmutation group or separate radionuclides and be changed if necessary, Under no conditions they must exceed the rated parameters of the accelerator complex.

It is quite obvious that to define the accelerator complex parameters firstly computer simulations must be performed and secondly there must be tests of separate units prototypes.

Calculations of specialists from LANL, IAERI, ITEP and other scientific centers have shown that designing of the high flux proton liner accelerator would require solution of the number of complicated technical problems. However the accumulated experience suggests possibilities for such problems solution. At least 2 such problems have the highest priority. The first problem is to create a powerful high frequency accelerator supply system of enhanced reliability and stability ruling out the necessity of remote control for the accelerator and its systems during shut down. Presently available high frequency supply systems (klystrons for 1Mwt) have insufficient continuous operation life time (about 8000 hours) .Thus the problem of improving the presently available high frequency supply systems and developing new ones with increased continuous operation life time is essential. As for the radiation situation on the accelerator structures during shut down it should be noted that it would be defined by the number of protons hitting the accelerator walls due to some deviations in the beam collimation system. The necessity to provide the linear accelerator high reliability and radiation safety during shut down makes us formulate the following requirement:

- The accelerator design and its supporting systems must ensure reliable continuous operation of the accelerator complex with the given efficiency of the operating time and radiation safety during its shut down for repair and maintenance.

Further calculations will show if this requirement will be left unrealized it wouid result in considerable losses in cost efficiency.

The junction unit of a accelerator-target is a very important part. Its aim is transport the proton beam from the accelerator to the target, A number of systems must be provided to ensure necessary conditions of the beam transporting. Namely there must be provided:

- permanent magnets for the beam bending at the right angle to the horizontal axis to protect the accelerator structures and systems against penetrating radiation from the target and blanket.

- quadropole lenses to receive a proton beam of the necessary size and its achromatization during bending at the right angle

– special appreture-radiation collimators to intercept protons with large horizontal coordinates

- devices for scanning the target surface to ensure averaging of the intensity in one unit of the target volume. The scanning system can be of no need if the neutron beam is split into several beams and there is sumultaneous irradiation of several targets

- vacuum systems with continuous oil free pumping provided with traps for heavy metal and noble gases emitted by the target and thus ensuring the necessary quality of the proton beam.

Even brief enumeration of supporting system for the junction unit stimulates serious attention to its designing, The preliminary studies of the operation modes of this block carried out at ITEP allow us to formulate the following requirements to its design and supporting systems,

To provide reliable operation of the junction unit the following parameters must be defined:

- the number of targets
- correspondence between the proton beam size and the irradiated target surface .
- vacuum system and gas traps capacity
- values of magnetic field strength and its homogeneity, possible limits of their changing by bending magnets, quadropole lenses, scanning or splitting magnets as well as their temperature ranges, possible fluence of the damaging radiation and the distance between the magnet system and the target.

It must be noted that for certain fission products and actinides no rated value of the proton current of 0.25÷0.3 A is required. It means that in this case no system of beams addition must be provided in the initial part of the accelerator and no splitting system in the junction unit. correspondingly. This made of the accelerator complex operation must be investigated further. During development of the electronuclear reactor in 1980-1985 ITEP specialists studies the performances of the supporting systems in the junction unit if proton energy is 1Gev and current equals 0.3A. E.g. according to our estimation the strength of the bending magnets field must be about 15 kOe and effective length must be 6m. For the beam achromatization system these parameters must be accordingly 0.5 kOe/sm and 0,8 m. Appretures of all magnet elements must be calculated taking into consideration the permissible level of the beam losses which makes up about 10^{-7} The beam size in this case is taken to be ~ 5 sm and homogeneity of irradiation is ± 10%. These values must be corrected for energy of 1.6 Gev defined for the accelerator complex by the later studies.

It was said about that at ITEP we considered the possibility of the proton beam scanning to ensure maximum beam intensity on the target and consequently to provide maximum density of the neutron flux in the blanket. However this approach requires further investigations. At the same time the version with the main proton beam splitting to irradiate several targets must also be considered. The results of these comparative investigations would help to design the target (or targets]. But even now some preliminary considerations about the target design and materials selection can be formulated. One of the problems we face in the target designing consists in separation of the vacuum chamber of the junction unit and the target volume. In order to maintain the proton beam intensity and to rule out remotedly controlled manipulations for replacement of the removable separating window it is desirable not to separate these volumes, To a certain extend this consideration influences the target materials selection and its state (solid or liquid]. For any version of the target the problem of its permissible intensity arises as well as the problem of receiving the maximal amount of primary neutrons for one proton, Our investigations show that a liquid target employing lead-bismuth alloys is more preferable because of good nuclear-physical properties for primary neutron generation and large experience accumulated in our country. But polonium-210 formation is a drawback of the material selected. Studies of the features of our target with lead-bisthmus alloys enable us to formulate the following preliminary requirements:

- To design and modes of the target exploitation must ensure the required level of primary neutrons generation and if necessary ensure the maximum level.

There must be a reliable heat and radiation removal from the target as well as residual heat removal. It necessary target material regeneration (purification) must be provided, The target must be reliably separated from the blanket and -other industrial premises and be provided with an appropriate biological shielding.

As for the blanket design at the present stage of our investigations ITEP specialists selected a heavy water blanket with thermal spectrum in accordance with the LANL specialists recommendations. For the target material we take salt $LiF-BeF_2$ containing nuclides for transmutation and this is also analagoas to the LANL recommendations. Preliminary requirements to the accelerator complex blanket are as follows:

The design and exploitation mode of the blanket must provide:

- the required rate of NT of HLW with minimal wastes loading into the blanket
- possibility to transform HLW into useful radionuclides
- possibility of the neutron flux multi purpose use including thermal and electrical energy production

reliable sealing of the structures and ensuring absence of radiation spread out side building in case of an emergency

means for the blanket remote diagnostics and maintenance

use of radiation resistive materials for the structures

- possibility to replace certain blanket units if necessary.

It must be noted that at the day stage of the concept study of the NT of HLW possibilities no requirements to the chemical technologies to be used for NT can be formulated.

Taking into account the fact that LANL specialists have also formulated a number of requirements to the NT processes and installations we strongly believe that there are good reasons for joining an effect and collaborating in this field.

5. BRIEF CONCLUSIONS.

1. The presented requirements to the accelerator complex for nuclear transmutation of HLW are preliminary and must be specified at further stages.
2. The requirements indicated must be taken into consideration in the accelerator complex design project.
3. After specifying the requirements presented can be used in the technical regulative documentation for designing and exploitation of accelerator complexes for NT of HLW.

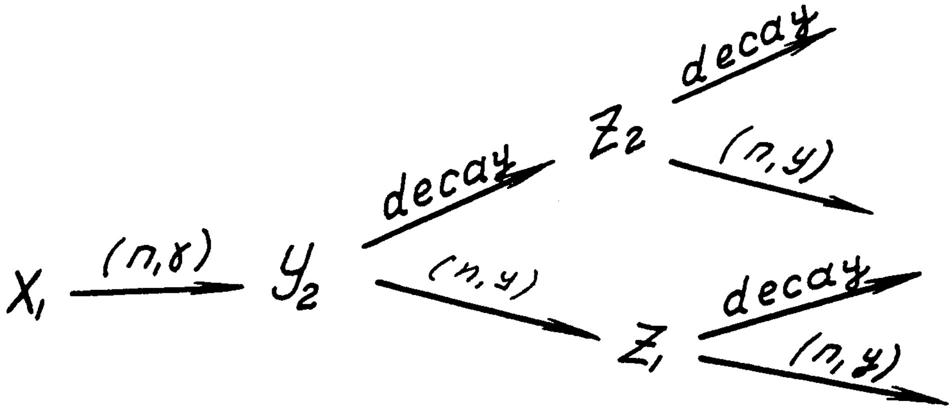

Fig. 1.