Journal of the Korean Nuclear Society
Volume 16, Number 4, December 1984

《Review》

Plutonium Recycle Strategy*

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(Received October 11, 1984)

I resingned PNC (Power Reactor & Nuclear Fuel Development Corp.) two years ago but still I am trying to contribute for the sound development of nuclear energy as a member of industrial group, so that I have been unacquainted with detailed technologies. However, I am repeating queations in my mind so often, what I had made for the development of Japanese nuclear power, what is the nuclear fuel, and so on.

The nuclear power is not only a means to generate cheaper electricity but also an important measure to improve national economy, to promote science and technology, and to create new employments in the advanced industries, and to say furthermore, nuclear technology is a most important key issue to the world peace and welfare. I believe.

There are only very limited natural energy resources both in your country and Japan. If we use electric power generated with petroleum, about 7,000 Yen in 10,000 Yen that is paid as its charge from us are to be flooded-out to abroad as fuel cost. All the equipments and materials needed for construction of fossile-fired power station are domestically made in our country. But here in your country, you may have to pay more Wons to import oil as well as some components.

Main fraction of nuclear power cost is capital, that is necessary to recover huge investments for construction of power station. One third of the power cost is fuel cost for which we have to depend upon foreign yellow cake, conversion and enrichment services. Most of discharged fuel is being sent to Europe.

Almost all the equipments and components are domestically obtailly obtained and fuel elements are commercially fabricated. So nearly 80% of money paid for nuclear power are circulating in Japanese economy to recall active industrial activities, such as cement industry, steel industry and manufacturing industries. Therefore it is natural for your government to have a policy to promote localization of nuclear power station. Financing of its investment has big influence on national economy depending upon whether the money is obtained in side the country or the money is obtained through loans from foreign countries. As the power generation is conducted by national corporation or authorized utility companies, the saving trends of a nation has close relationship to their own national economy.

There are very limited amounts of uranium resources in Japan, and at present no commercial uranium enrichment capacity. Centrifuge technique has been developed up to a stagn of construction of a demonstration plant, but only one-third of future requirement is to be produced

* 1984년 10월 11일 한국에너지연구소에서 강연한 내용중 발췌한 것임.

in our country, taking into accounts of world demand-supply situations, investments needed for development and commercial plant as well as possible business life which is largely affected by introduction of commercial FBRs, though no one can definitely estimate. New technique such as laser enrichment is being studied because of future forecast of elongated LWRs days than formerly expected and availability of natural uranium in the world.

Once the FBR and its fuel cycling is technically and economically developed, electric power needed for a nation could be produced by brain and sweats of the nation. It will moderate the pressure of international conflictions, mainly caused by hasty export of industrial products or in some case export of weapons, because the nation could reduce oil and coal import dependency.

This is why I said earlier that the development of nuclear cnergy is key issue for world peace.

Then is it necessary and feasible for each nation that more and more LWRs are introduced and early development and commercialization of FBR are promoted? It is not so simple. There are many limiting factors.

The experimental fast breeder reactor JOYO has been operated for these years. A prototype FBR is under construction. Design studies on demonstration reactor have been done both for loop-type and tank-type. Several fuel pins discharged from JOYO with 50,000 MWD/T at maximum were experimentally reprocessed, and next test operations using high burn-up fuel such as 120,000 MWD/T are prepared. Recovered plutonium is converted and fabricated to a test pin to be charged again in JOYO. Thus the FBR fuel cycling is being closed even though its scale is very limited.

However, we will encounter more difficulties to develop full scale fuel cycling in commercial FBRs. FBR days including commercial reactors and fuel cycle facilities will be reached after a few decades. Formerly an optimistic opinion that commercialization of FBR system is to be done around 2000 was believed, but recently it is said to be around 2010, 2020 or 2030 and later.

In order to shift from LWR system to FBR system, spent LWR fuels must be reprocessed. Reprocessing is technically feasible as PNC demonstrated, but reliability and economy of reprocessing are to be demonstrated in the future. Owing to foreseen various difficulties to develop commercial reprocessing plants, one cannot be so optimistic. Many talks are being discussed as following, what is the plutonium balance after completion of second reprocessing plant? How much spent fuels are to be stored when the second reprocessing plant is duely constructed or with some delay? What are the adverse factors for the second reprocessing plant, from standpoints of social, economical, or international influences?

1. Plutonium is an alive material

Usually formation of Pu-239 from U-238, through Np-239 and then isotopic change to Pu-240, 241, and 242 in the reactor core are considered. Amount of total fissile plutonium is used for approximate calculation of nuclear fuel balance and for study on fuel cycle strategy. It is true, but is not sufficient for actual planning of fuel cycle.

U-238 is converted to U-237 as (n, 2n) reaction with minor probability. Then Np-237 changes to Np-238 and Pu-238. Part of Np-237 converts to Np-236 as (n, 2n) reaction and then changes to Pu-236.

Pu-241 decays to Am-241 with a half life of 13.5 years. various nuclear reactions produce Am-243, Cm-244, etc. as trans-plutonium elements. Those trans-uran elements have peculiar radiological properties that must be considered in order for safe handling. Alpha decays and beta decays of shorter half-life must be carefully considered both the reaction themselves and their daughter isotopes. Minor content of U-232 cannot be neglected because it decays rapidly through various elements some of which emits hard gamma orhard X-ray, and requires shielding and handling difficulties.

Content of Pu-241 in plutonium recovered from LWR spent fuel of 30,000 MWD/T after one year's cooling is 10~12%. Fissile value is lost with a half-life of 13.5yr. Daughter isotope of Am-241 causes handling difficulties both from standpoint of safe operation and of reactor physics, especially in FBR fuel design and fabrication which has higher plutonium enrichment.

Anyway plutonium is not a stable element but alive material.

This is an experience obtained about fifteen years ago. We conducted a series of measurement of nuclear reactivity of plutonium fuel rods using light water moderated critical facility in JAERI. As for the usual low enriched uranium fuel experiments, rods are positioned to a specified lattice prior to measurement, and moderator water is introduced up to specified water-levl, and then neutron source such as Ra-ße is inserted to the core. Neutron multiplication factor is counted, experiment is repeated again and again with increased water level. Finally the system becomes and go over criticality. Thus a relative nuclear reactivity of fuel rod, or rods, or assemblies is calculated.

Number of MOX fuel rods consist of natural UO_2 and PuO_2 of around 2% enrichment was increased step by step similarly to uranium rods experiments. One day when more than one hundreds MOX fuel rods were used for experiment, the core system brought into criticality without insertion of neutron source. As the facility is designed guite safely, the phenomenon was not a incident but just a surprise. It was natural from physical standpoint. Pu-240 and Pu-242 emit neutron as the result of spontaneous fission, which acted as a neutron source of the experiment. Plutonium is alive.

Recently such an experience was being obtained. Fissile content in plutonium when we started research works in 1966, was about 9%. The content is getting lower and lower. Recent value is near 65%. So the radiological precautions were mainly alpha protection in early stage, but neutron protection emitted from Pu-240 and 242, and gamma protection owing to Am-241 daughter, as well as to minor contents of Pu-238 and 236 became necessary to avoid undue exposure of operators. PNC applies the direct de-nitration method by micro-wave heating of mixed plutonium-uranium nitrate solution to obtain mixed oxide powder. Am-241 build-up over several thousand ppm, and no purification of solution was applied in this process, so that personal lead shielding apron became necessary. Such events had been fully anticipated and new MOX fuel fabrication facility is under construction in which almost all operations are automated and maintenace of equipments is done after removing plutonium remotely from that point of glove-box.

There are several question;

Is it possible to fabricate MOX fuel at a reasonable cost using such an expensive facility? Is the facility operated with reasonable plant factor?

Is it better way to have another purification facility to remove trans plutonium elements and their daughters than to have such sophisticated fabrication facility?

Purification of stored solution is fairly expensive, and if one has to purify stored plutonium oxide or mixed oxide powder, much expensive facility is necessary.

A report from Belgo-nucleaire shows practical maximum permissible storage periods for each fuel form. This is a simple expression applicable to their fabrication facility in which direct glove-box operations are still applied.

As you understand now, plutonium is indeed alive material. It is incorrect to express definite figure as MOX fuel fabrication cost and it is not wise way to establish nuclear fuel cycle strategy based on such unfixed figures.

I said that discussions on nuclear fuel balance and fuel cycle strategy are repeated in former section. We had similar discussions around 1970 when Japanese economy was rapidly expanding and LWR became to be thought as a prime measure in future nuclear power.

How much natural uranium and enrichment service are available for Japan?

Is it possible to develop advanced power reactor systems by our own efforts?

Since then Japanese nuclear industry made a successful progress, so that more precise analysis is made and more options are available.

In these years we have been operated a fairly well equipped plutonium facility since 1966. Fabrication studies, fuel characteristics studies were conducted. As plutonium has quite different reactor physics, a series of critical experiments was carried out as a joint study with JAERI. We sent engineers to ANL and Fermi to study about FBR.

Now we have experiences to recover plutonium born in Japanese LWRs, to handle higher plutonium isotopes and to fabricate MOX fuel more than 70 tons. Problems concerning safety control, safeguards, wastes management and economic analysis are fairly well understood. On the other hand there are many questions.

2. Management of Spent Fuel

Plutonium is alive in spent fuel. It had worked in situ in the reactor core where it was born. About one third of burn-up of discharged fuel was contribution of plutonium. Pu-241 stopped nuclear reactions both fission and conversion but still decays to Am-241. As its decay half-life is 13.5 years, total fissile plutonium isotopes decreases from 65% to 60% or less in ten years for spent fuel with burn-up of 30,000 MWD/TU. Pu-238 and 236 are minor components that are valueless because of their non-fissile property. But decay daughters contaminate uranium radiologically. So long range storage of spent fuel must be considered from view point of such nuclear reactions as well as economic disadvantage caused by a fact that valuable material is to be stored, and that storage itself is quite costly.

However in reality, there are not sufficient capacity of reprocessing in the world, stable operation of a reprocessing plant is still a problem to be improved, and reported reprocessing cost is quite expensive. In United States that has maximum nuclear power generation capacity, there are no commercial reprocessing plants and no expectation for coming plants. There are plentiful energy resources such as coal, gas and petroleum, and vast deserts. Nuclear power becomes unattractive under these economic conditions, so that many of plans for nuclear power station are being cancelled, while studies on disposal of spent fuel assemblies are under way.

Japanese utility companies send their spent fuel for reprocessing to Europe as well as to Tokai. They are preparing to construct a large scale commercial reprocessing plant. Experiences in Tokai plant, informations from advanced European and American reprocessing industry and results of R&D in Japan sponsored by

Government are to be combined for design and construction. Many procedures including public acceptance are necessary. and high plant operation factor could hardly obtained in its earlier stage of operation. Anyway world's experience on LWR fuel reprocessing is still insufficient to be optimistic. Improvement are to be obtained in coming plants, but definite conclusion on reprocessing economy is still difficult.

There are slight change of views on reprocessing in recent Japan. Originally a big capacity such as 1,200 tons or 1,500 tons was planned for the second reprocessing plant keeping pace with accumulation of spent fuel.

The capacity is reduced to a level enough for demonstration of improved technology, and plutonium obtained is to be used demonstration EBR and early commercial FBR as well as well as possible commereial ATR. Plutonium recycling in LWR is to be done in order to avoid excess storage of the sensitive material. The second reprocessing plant is designed to be able to accommodate future construction of additional capacity.

Therefore the intermediate storage of spent fuel is one problem to be considered. Increase of storage capacity at reactor site, expanded storage pool at the second reprocessing plant and dry cask storage are being investigated.

Intermediate or long-term storage of spent fuel is also studied in. U.S. These informations will be useful for your country.

Plutonium recycling in LWRs is justified for countries such as France and Japan where large scale reprocessing plant exists or is to be constructed as a fixed policy, from view points that excess stockpiling of plutonium is not desirable technically, economically and politically. It will be an important measure for plutonium management and a measure to recover some portion of investment for reprocessing plant. Also it will be a necessary step to create industrial basis for MOX fuel fabrication in future FBR days. However, plutonium recycling may not be justified economically for a country where no fixed program for FBR development and no plan of construction of reprccessing plant are yet decided.

3. Towards the Future

Up-3 in France, THORP in U.K., WA-350 in West Germany and maybe JNFS plant in Japan as well as BRET for reprocessing study of spent fuel FFTF fuel in U.SA. will be constructed and put into operation in these ten years. Japanese test facility for FBR fuel reprocessing will also be in construction stage

All these reprocessing plants adopt PUREX process as their fundamental processing, and more reliable and advanced equipments and materials will be used. Sophisticated design for maintenance system will be applied. Robot application will be done, and remote replacement of failed equipment in exchangeable rack will be tested. Once these plants demonstrate their applicability and experiences to reprocess higher burn-up fuels are accumulated in the world, reliability and cost estimation for the third generation reprocessing plant will be precisely obtained.

Experiences on solidification of high active waste will be obtained in several plants. Disposal of HAW is hardly realized in these ten years, but social acceptance will be much better obtained.

MOX fuel fabrication experiences will also be increased, and uncertainty on MOX fuel fabrication cost mainly come from processing and disposal of TRU waste is expected to be solved.

The latest ANS conference held in Wyoming in which reprocessing and waste treatment were listed as its topics was a symbolic event that the ambiguous tunnel years on fuel cycling in the last decade is being passed through.

As the Republic of Korea is now entering into advanced industrial countries, it is advisable to strength its policy on nuclear power keeping eyes on recent tendencies in the world.

Localization of A/E and components for nuclear power station would be a prime target. Uranium fuel fabrication could be commercialized as the demand grows up to industrial scale.

Spent fuel must be stored as an important energy resource mine in the future, as your country is not blessed with natural resources as our country. Plutonium is alive material as I repeated, but seems to me that pre-mature decision on when spent fuels are to be reprocessed, and how the plutonium is to be utilized based upon too much earlier observations, is not advantageous.

Plutonium technology is a key for future nuclear engineering. It has special delicate properties physically, chemicallly, metallurgically, and radiologically and it has a big influence on nuclear econmy.

Therefore it will be necessary for you to start various studies in the future in order to obtain industrial basis. However plutonium is sensible material in view of the international policy on non-proliferation of nuclear explosion. So that the planning and executing is desired to be done carefully and evolutionally.

Large scale reprocessing of spent fuel and fuel recycling which would be required in this country beyond the turning of this century, is hopefully conducted as an international venture This is just my personal opinion.