NONPROLIFERATION CHARACTERISTICS OF ADVANCED FUEL CYCLE CONCEPTS*

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ABSTRACT

The purpose of this study is to comment on the proliferation characteristic profiles of some of the proposed fuel cycle alternatives to help ensure that nonproliferation concerns are introduced into the early stages of a fuel cycle concept development program, and to perhaps aid in the more effective implementation of the international nonproliferation regime initiatives and safeguards methods and systems. Alternative cycle concepts proposed by several countries involve the recycle of spent fuel without the separation of plutonium from uranium and fission products.

INTRODUCTION

Several countries have proposed the recycling of spent fuel without the separation of plutonium from uranium and fission products. The concepts are alternatives to either direct long-term storage or the PUREX® reprocessing of spent fuels. Some of the fuel cycles being investigated include the dry-recycle processes such as the direct use of reconfigured PWR spent fuel assemblies into CANDU reactors (DUPIC), and the thorium/uranium cycle systems. The nonproliferation advantages usually associated with these non-separation processes are: (a) the highly radioactive spent fuel presents a barrier to the physical diversion of the nuclear material from the fuel cycle; (b) the need to dissolve and chemically separate the plutonium from the uranium and fission products, and (c) the spent fuel isotopic quality of the plutonium vector is further degraded. Although high radiation levels and the need for reprocessing may be perceived as a barrier to terrorists or other sub-national groups, international proliferation concerns are addressed primarily by the material accountancy and verification activities which are the international safeguards measures of fundamental importance, with containment and surveillance as important complementary

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measures. Consequently, the non-separation fuel cycle concepts have to be evaluated on the basis of the impact that the fuel cycle processes may have on nuclear materials accountancy. Safeguards R&D would have to address the implementation of advanced non-destructive assay and accountancy methods for dry-processing systems having concept specific holdup characteristics (measurable and non-measurable). The objectives of this paper are: (1) to profile the proliferation characteristics of some of the fuel cycle concepts; (2) to suggest that the non-proliferation concerns be introduced into the early stages of a development program, and (3) to perhaps aid in the more effective implementation of international non-proliferation initiatives and safeguards methods.

CANDU/PWR CYCLE

The residual reactivity in the spent fuel of a PWR cycle can be used to extend the fuel burnup capability in the high neutron economy of heavy water reactors. The direct use of spent PWR fuel in CANDU reactor (DUPIC) has the potential of reducing the natural uranium resource requirements, and of reducing the overall spent fuel arisings by refabricating spent PWR fuel into CANDU reactor fuel bundles for power generation.[1] The advantages of the DUPIC cycle are outlined as: (1) no wet-processing is required; (2) the geological disposal requirements of the spent PWR and CANDU fuels are greatly reduced; and (3) the natural uranium requirements for the CANDU phase of the fuel cycle would also be greatly reduced.

The DUPIC fuel cycle concept utilizes dry-bulk-processing techniques. The dry process involves the mechanical dismantling, segmenting, and decladding of the PWR fuel elements, and the grinding of the U/PuO_2 spent fuel into powder. The spent fuel oxide powder is then subjected to cycles of oxidation/reduction processes, sintered into pellets and configured into CANDU fuel bundles. The process removes a high fraction of gaseous and volatile fission products. The remaining fission products with high gamma and neutron radiation levels necessitate that refabrication and fuel assembly handling be shielded and automated. Although the gamma radiation and neutron emission levels could be perceived as a layered barrier to deter sub-national theft or diversion, the radioactivity does not present a layered barrier to the national diversion and the international level of proliferation concerns. The proliferation resistance characteristics address the barriers to accessing the material, and the barriers to converting the nuclear material for uses other than for power generation.

International proliferation concerns require that stringent safeguards in nuclear material for uses other than for power generation. International proliferation concerns require that stringent safeguards in nuclear material accountancy and validation be implemented throughout the dry-bulk process. The radiation barriers could compromise the efficacy of the accountancy and validation measures. With respect to barriers in converting the nuclear material to non-fuel uses, the softer neutron spectrum in the CANDU reactors generates a plutonium isotopic vector with greater concentrations of the even plutonium isotopes (Pu-240 and -242) than in the initial PWR spent fuel feed. The fresh fuel DUPIC plutonium isotopic composition for a 35 GWD/T PWR power operation, is transmuted into a plutonium isotopic composition for a 16 GWD/T DUPIC once-through power cycles. The specific neutron emission rates of the even-plutonium isotopes (equivalent to Pu-240) in the DUPIC fuel is found to be increased by factors of two or more than in the initial PWR or CANDU spent fuel arisings, maintaining the plutonium more unattractive for non-fuel use.

In the dry-bulk process, the nuclear material forms, inventory, and flow are such that current non-destructive assay (NDA) methods may be limited. Research and development would have to address specific measurement methods and validation standards to directly assay the nuclear materials in the two major waste streams from the mechanical dismantling, segmenting, and decladding processes of PWR fuel assemblies and from the oxidation/reduction, sintering/pelletizing process. Dry-bulk powder processes have characteristically high holdup inventories and excessive nuclear material holdup could be expected in the DUPIC dry-bulk-processing plant. The early stages of a feasibility exploration program would have to include early design considerations which address the following: (a) proliferation characteristics based on the radiation level of the fuel assemblies as being within the sub-national level threat or as an international level of proliferation concern, (b) geological repositories would still be needed for the once through DUPIC fuels, and (c) impact on the IAEA materials accountability measurement methods for establishing dry-process holdup and recoverability of the plutonium/uranium mixture.

Impact on Geologic Repositories

The impact on the repository requirements for long-term storage of the once-through DUPIC spent fuel was estimated for a ten year cooling period in interim storage. The nominal radiation and heat emission rates per ton of heavy metal (THM) were compared with the

emission rates of a reference LWR-UO_2 once-through cycle (35GWD/THM) and the CANDU fuel cycle (7 GWD/THM). The scoping estimates indicate that a minimal impact is to be expected for the DUPIC cycle on the repository design specifications for a reference LWR-UO_2 once-through fuel cycle. Spent fuel storage area requirements and inventory verification measurement methods are not significantly affected.

THORIUM-URANIUM FUEL CYCLES

The thorium-uranium fuel cycle is being pursued on a limited scale by several countries. The primary incentive for the thorium-uranium fuel cycle would be to utilize indigenous nuclear material resources in conventional commercial thermal and fast reactor power systems. The exploitation of thorium resources is essentially a long-term, large-scale energy-generation option.[2,3] The U-233 produced in the Th-U cycle is as fissionable and perhaps as radiotoxic as Pu-239. Although systems have been proposed in which U-233 is denatured with U-238, even the denatured Th-U/U-233 introduced non-proliferation concerns. The suggested cycles include U-233/U enrichment levels ranging from three to twelve percent or more.

The thorium-uranium fuel cycles have certain nuclear characteristic features, which persist through all stages of the fuel cycle process and consequently may strongly influence the design of IAEA safeguards. Two fundamental invariant characteristics in the denatured uranium-thorium fuel cycle are: (1) chemical reprocessing is a necessary phase of the fuel cycle; and (2) the U-233 and the plutonium isotopes are in combination throughout the back end of the cycle. The separation of the uranium-thorium and plutonium is necessary for reactor systems in a symbiotic power complex operating at high conversion ratios with only denatured uranium (low enrichment in U-235 and U-233) being used in the front end of the cycle. Studies in EMIS processes (electromagnetic isotope separations) have indicated that the separation of U-233 is greatly facilitated in comparison to the isotopic separation of U-235 in the low enrichment U-235/U cycle.[4] The estimates indicate that the effort level to enrich U-233/U fuels, can be reduced by factors of 3 to 20 in comparison to the re-enrichment of 3% U-235/U. The ease of the isotopic separation is a consequence of the mass difference of five between U-233 and U-238 as compared to three for the U-235 and U-238, the higher concentration levels of the U-233/U fuel, and the lowered inventory fuel requirements to achieve significant-quantity mass levels.

The anticipated impacts on safeguards of the thorium-uranium fuel cycles are speculative, since sustaining fuel management strategies have not yet been detailed. The source of the U-233 for startup, the disposition of the plutonium generated in the denatured fuels because of the presence of U-238, and the precise composition of the U-233/U fuel should be evaluated as the fuel cycles are developed further. The uranium isotopes for moderately high burnup can be nominally in w/o 0.1/ 81/ 15/ 3/ 0.2 in U-232/ 233/ 234/ 235/ 236, respectively. In the thorium blanket, the isotopics could have nominally 0.2/ 94/ 05/ 0.5/ 0.03 in w/o for the corresponding vector. For high burnup in the thorium-uranium cycle, the plutonium isotopic concentrations should approach the level in LWR fuels for 50GWD/T of about 3.6/ 47/ 28/ 11/ 10.5 w/o in Pu-238/ -239/ -240/ -241/ and -242, respectively. The safeguards assessment of the U-233/U/Th fuel cycles would involve the layout of symbiotic systems to establish the somewhat unique aspects of the fuel: (a) feed sources of U-233 would be necessary since the denatured cycles may not be self-sufficient in flow and inventory of fissile material; (b) the impact on safeguards relating to the differences in the enrichment properties and capacity requirements between U-233, and U-235; (c) the impact on IAEA safeguards methods resulting form the radioactivity of the daughter products of the U-232; and (d) proliferation concerns be addressed at the front-end of the fuel cycle. The safeguards problems introduced into the nuclear material accountability methods by the thorium-uranium cycle will require research and development programs in implementing advanced chemical analytical measures and NDA techniques.

Impact on Repository Requirements

The nominal specific radiation and heat emission rates for the seed fuel cycles are not expected to differ significantly from the emission rates of the LWR-UO_2 once-through spent fuel standard. The blanket fuel cycle designs in some concepts undergo much higher levels of burnup compared to the Th/U seed and LWR-UO_2 fuel cycles. Consequently, the decay heat and the neutron emission rates for the blanket fuel could be expected to range by a factor of 2 to 3 higher than the decay heat emissions and by a factor of 5 to 6 higher for neutron emission rates, than the Th/U seed fuel and the LWR-UO_2 reference fuel cycle. Geologic repository designs should be phased into the early design stage of Th/U alternate fuel cycle concepts.

EXCESS PLUTONIUM DISPOSITION CYCLES

The disposition of excess weapons plutonium from disarmament programs has been explored in many studies and by many countries in the past few years. There appears to be a general consensus and commitment for the continued study of two major options: use in reactors, and immobilization.[5] The reactor alternative involves the use of MOX fuel as a fuel source for commercial reactors (LWR's, VVER's, and Fast Reactors).[6] The immobilization alternative involves the vitrification of plutonium in a matrix log inserted within a canister containing radioactive material (can-in-canister). The consensus also included the time lines: the plutonium disposition program is to be initiated in approximately 10 years, and the program should be completed in 25 to 30 years thereafter.

Thermal Reactor Fuel Cycle (LWR, VVER)

In the case of the MOX fuel burning in thermal reactors, the once-through fuel cycle operation degrades the weapons-grade plutonium into a form that is as unattractive and inaccessible for weapons use as that of plutonium contained in the spent fuel from current commercial reactors (Spent Fuel Standard). The use of thermal reactors would transmute the weapons plutonium into spent fuel plutonium isotopic nominal concentrations in weight percent of about 0.13, 56, 28, 13, 3 for Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, respectively for a once-through 40-45 GWD/T burnup cycle. The specific neutron emission rate from the even-plutonium isotopes, "equivalent" to Pu-240, is expected to be comparable for the reference spent fuel standard (SFS) and the once-through fuel cycle of the LWR/VVER MOX loading.

Fast Reactor Fuel Cycle

The burning of weapons plutonium in fast reactors with conversion ratios of less than unity results in a plutonium composition vector, for the once-through cycle (open cycle) in nominal range of 0.1, 84, 14, 1.1, and 0.1 weight percent for Pu-238,m Pu-239, Pu-240, Pu-241, and Pu-242, respectively. The isotopic degradation does not differ much from the initial weapons-grade plutonium. The plutonium isotopics would approach the LWR spent fuel standard concentration levels, if an equilibrium feed and discharge fuel recycle mode of operation is utilized in the fast fuel cycle or in a once-through irradiation exceeding 150 GDW/T. However, the equilibrated fuel cycles would require higher burnups equivalent to

some 4 to 5 recycles which would then exceed the time line for completing the disposition program.

Impact on Repository Requirements

The representative nominal specific radiation and heat emission rates for the weapons-grade (WG) plutonium once-through MOX spent fuel are compared with the LWR-UO_2 once-through cycle and the first RG Pu once-through recycle MOX fuel. The scoping estimates seem to indicate that WG Pu MOX spent fuel should have minimal impact on repository design specifications for the representative spent fuel standard of the LWR-UO_2 once-through cycle. The WG Pu MOX spent fuel can be stored in a UO_2 spent fuel repository with clad temperatures of the WG Pu MOX being well within the UO_2 limits.

The reason for this is that the actinides (Pu-238, Am-241, and Cm-244) which contribute to the heat load must go through the Pu-241 chain. The UO_2 and WG Pu have no significant initial concentration of Pu-241, and consequently the higher actinides build-up in like manner for the once-through UO_2 and the WG Pu MOX fuels. In contrast the recycled RG Pu MOX have a decidedly higher initial concentration of Pu-241, and the build-up chain to the higher actinides (Pu -238, Am-241, and Cm-244) is very pronounced.

SAFEGUARDS MEASURES

The need for advanced safeguards development in direct plutonium accountability measures exists in all of the proposed disposition options. NDA and DA systems development for the dry-bulk processes should be advanced: (a) to enhance the transparency of the weapons-plutonium flows and inventories; (b) to improve material accountability, (c) to directly establish isotopic compositions; and (d) to ensure that the weapons material are indeed unattractive, inaccessible, and irreversible for uses other than the nuclear power fuel cycle. Advanced NDA and DA methods for direct plutonium accountability, being developed in many analytical chemistry laboratories, should be explored for implementation on an engineering scale in on-line processing operations.

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