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Abstract

The Great East Japan Earthquake and Tsunami in 2011 led to the accident at the Fukushima Daiichi Nuclear Power Plant (1F) and subsequent meltdown of the reactor cores of Units 1, 2, and 3. The Japanese project on material accountancy technology development for fuel debris of Units 1-3 of 1F has been implemented under the roadmap, "Mid-and-long-Term Roadmap towards the Decommissioning of Fukushima Daiichi Nuclear Power Units 1-4" with recovery of the fuel debris starting in 2020. Japan Atomic Energy Agency (JAEA) and United States Department of Energy / National Nuclear Security Administration (DOE/NNSA) have agreed to collaborate to investigate past experience on material control at TMI-2 and ChNPP. This paper describes the material accountancy processes employed on the damaged TMI and ChNPP nuclear materials. This paper also explores how the material forms retrieved from these accidents could be placed under International Atomic Energy Agency (IAEA) safeguards, and identifies material characteristics that may be encountered at Fukushima Daiichi that would impede the application of safeguards.

Study Methodology

The Chernobyl Nuclear Power Plant (ChNPP) portion of this study is based on first-hand experience of U.S. laboratory experts, IAEA staff, and Chernobyl safeguards experts who evaluated new safeguards approaches that could be used at ChNPP, and literature searches of studies and reports written in the aftermath of the Chernobyl accident. The U.S. team worked directly with experts from Ukraine to devise measurement systems that could be used by Ukraine to make declarations to meet their obligations under their Comprehensive Safeguards Agreement (CSA) with the IAEA, and with experts from the IAEA to devise containment and surveillance systems that could be installed at ChNPP to confirm that all nuclear material remained in peaceful uses.[1,2,3]

The Three Mile Island (TMI) Unit 2 (TMI-2) portion of this study is also based on the first-hand experiences of the technical experts who were responsible for the removal and accountancy of nuclear material from the damaged TMI-2 reactor. While not under international safeguards obligations, U.S. domestic law required a nuclear material accountancy approach for the damaged TMI-2 fuel. This approach centered on the comparison of the calculated special nuclear material (SNM) in the core in TMI-2 at the time of the accident, to estimates of the small amount of SNM left in the core after recovery operations were completed, and measurements on the shipped materials themselves.

Background information about both the ChNPP and TMI-2 accidents is not included in this paper. This paper focuses instead on specific activities related to Nuclear Material Accountancy (NMA) after the ChNPP and TMI-2 accidents. In some instances, it may not have been possible or cost-effective to perform NMA activities to provide verification of nuclear material amounts and locations to desired levels of certainty, specifically at ChNPP. Furthermore, some of the NMA practices may not have practical bearing on the Fukushima situation. These practices are examples of process development and capture a comprehensive list of possibilities that may be applicable to Fukushima.

ChNPP

At the time of the ChNPP accident (1986), Ukraine was still part of the Soviet Union. As such, the nuclear material after the accident was subject to domestic accountancy requirements, but not international safeguards criteria. After Ukraine gained its independence in 1991, and ratified the Non-Proliferation Treaty (NPT) in 1994, nuclear material became subject to IAEA verification. This effectively created two distinct periods of post-accident nuclear material management at ChNPP; pre-IAEA material accountancy, and IAEA safeguards. While there are significant technical differences between the accidents at ChNPP and Fukushima Daiichi (1F), there are potential lessons learned with respect to the material accountancy methodology under IAEA safeguards for a facility that can be described as "destroyed by beyond design basis accident," as well as the practical implementation of safeguards measures under severe accident conditions.

After the accident, ChNPP personnel began activities to determine the status of the damaged core.[3,4] These initial efforts were focused on ascertaining the radiological conditions around unit 4 as well as the status and location of nuclear material. The importance of the latter was primarily to examine if there were criticality issues wherever the fuel was located. In effect, these activities became the first material accountancy estimates after the accident.

These post-accident efforts started with simple visual observations and developed into later empirical measurements of heat production and isotopic surveys. The visual examinations focused on categorizing Fuel Containing Masses (FCM) by appearance and location. Initially, the visual surveys resulted in 3 broad FCM categories:

1. UO_2 fuel pellets or partial assemblies (fuel fragments)
2. Dispersed dust and aerosol
3. Melted Lava-Like FCM (LLFCM)

In addition to identifying the category and location, observers attempted to quantify the mass of nuclear material through volumetric estimation. There were several potential sources of error associated with this approach; consistency of the observer, recollected knowledge of the volume containing the FCMs, debris covering FCM, and density variations of the FCMs just to name a few (Figure 1). Regardless, the observations provided the opportunity to begin classifying the nuclear material for both immediate safety analysis, but also for later assessment to confirm presence and any material changes that were occurring.

Figure 1. ChNPP personnel perform visual examinations in damaged areas of Unit 4 reactor to estimate FCM locations. Photo Credit: Chernobyl Center

After visual inspections were completed, a series of heat measurements were taken from 1986-1989. These measurements attempted to use heat probes to measure thermal fluxes as physically close as possible to the visually confirmed FCM. The time correlated heat flux data was compared against calculated heat generation capability of the nuclear fuel. Ultimately, there were several factors that limited the usefulness of this approach; 1) Radiological conditions in various parts of unit 4 prevented measurements that could isolate individual FCM accumulations. 2) Heat output from the nuclear material was eventually determined to be significantly less than heat output of equipment used during recovery operations. 3) Convective heat exchange between unit 4 and the remainder of the ChNPP units gave rise to a thermal background approximately equal to the calculated heat output of the unit 4 core. Some analysts[5] contend that this approach may still be viable if specific conditions can be met. These conditions include better and more uniform distribution of thermal sensors, complete control over any other heat-generating equipment, and more accurate information about the isotopic composition of the damaged core. To date, no further work has been done to estimate nuclear material mass at ChNPP via thermal methods.

A third method for evaluating material masses within unit 4 and ChNPP involved measuring the ^{137}Cs activity around the ChNPP site and FCM accumulations, and then comparing these values with the total calculated ^{137}Cs loading, and an experimentally determined ^{137}Cs retention in the damaged fuel. This approach only takes into account fuel that was destroyed and converted into a dust/aerosol, or melted into a LLFCM. Visual observations however showed that there were significant distributions of fuel fragments throughout unit 4, as well as outside the damaged reactor. Since much of the ejected material was buried without any attempt to quantify nuclear material mass, the ^{137}Cs balance method is viewed as having large uncertainties along with the visual and thermal approaches.

After ratifying the NPT, Ukraine submitted an updated Initial Inventory Declaration for Chernobyl Unit 4 to the IAEA in 1998. This declaration included fresh fuel, core fuel, and spent fuel. It took into account nuclear production and loss with estimated plutonium content and uranium burn up, respectively, as of the date of the accident. It did not take into account any losses of the nuclear material in the Shelter since the time of the accident and the whereabouts of that material.[2]

Between the period 1998 and early 2003, the IAEA and Ukraine implemented safeguards under the premise that detailed accountancy could not be performed at Unit 4 and that safeguards could be implemented by a combination of containment and surveillance measures (C/S) and the physical security system that surrounded the damaged reactor. So long as no SNM crossed the boundary of the protected area surrounding the Chernobyl reactors, the IAEA accepted that the material remained under safeguards. Implicit in this approach is that the risk of material being transported out of the protected zone was small given that there were few people working on the Chernobyl site and there was no large equipment that could be used to remove SNM from the damaged reactor. This status quo changed in early 2003.

In 2003, Ukraine and an international consortium comprising Battelle Memorial Institute, Bechtel, and Electricie de France (EDF) began an initiative to strengthen the existing Shelter structure because it was under the threat of collapsing, and construct a New Safe Confinement (NSC) to replace the old Shelter. This would ensure that the damaged reactor would be contained for years to come. Once this activity begins in earnest, there will be a large number of people and heavy machinery onsite. The increased level of activity, the number of people, and the availability of heavy machinery increased the risk that nuclear material could be removed clandestinely from the Shelter and from the site. The huge NSC will cover the old Shelter and will have cranes installed from its roof capable of removing the old deteriorating old Shelter roof which would give access to the damaged Unit 4 reactor hall and spent fuel ponds. These cranes could be used to clear up the rubble in this region and gain access to the 129 fairly intact spent fuel assemblies containing low-enriched uranium (LEU) and plutonium and 48 somewhat damaged fresh fuel assemblies containing LEU. The spent fuel may still be suitable for packaging in a cask for removal and eventual reprocessing. The LEU in the fresh fuel could have use feeding an enrichment plant or fueling a plutonium production reactor. Hence, one must worry about the capabilities of the NSC to remove the material that is most desirable from a diverter's viewpoint in the Shelter. The safeguards approach will dwell heavily on these last two problem areas. The consulting team has at this stage in the project studied these crucial challenges and described some preliminary paths for dealing with them.

The IAEA recognized that they needed to update the safeguards approach for Unit 4 with the impending construction of the NSC. In 2003 they took the lead and convened an international team to analyze the impact of the NSC construction on safeguards at Chernobyl Unit 4 and devise a response that would ensure that the safeguards approach would address changing circumstances and evolving risks. The team included staff from the Department of Safeguards Operations Division C, Section 2 (SGOC2); the Division of Technical Services (SGTS); the Division of Concepts and Planning (SGCP), the State Nuclear Regulatory Committee of Ukraine (SNRCU), ChNPP, and representatives from US national laboratories and Russia. The core team comprised 12 people, with other experts supporting various technical studies as required. The IAEA understood that it was important to establish a team with broad expertise in safeguards implementation, monitoring system design and equipment development; include experts from the SNRIU and ChNPP, be sufficiently large to share the expected workload, access to IAEA extra-budgetary funding.

The IAEA also understood the interrelationship between Ukraine's declaration and the IAEA's verification activities. Since the IAEA perceived that Unit 4 did not fit any of the facility categories in the IAEA Safeguards Criteria, Ukraine needed guidance on how to declare the facility, nuclear material and implement safeguards at Unit 4. Therefore, neither Ukraine nor the IAEA had justification and guidance to complete a design information questionnaire (DIQ), implementing nuclear material accountancy, and verifying declarations. All the issues related to implementing safeguards at Unit 4, from Ukraine developing its DIQ and declaration reporting basis to the IAEA safeguards approach needed study and clarification to proceed. The approach of using a joint team facilitated this process. It promoted the timely exchange of information between the IAEA and Ukraine, ensuring that information was quickly shared with key decision makers so that those responsible for designing the safeguards systems knew what was expected of them and the capabilities and constraints that would be imposed on any accounting or monitoring system.

The team met regularly over the course of the project, alternating meeting locations between IAEA Headquarters in Vienna and SNRIU offices in Kiev, Ukraine. By alternating meeting locations, each side was able to invite local staff to participate in the meetings without incurring additional travel costs. The regular meetings kept the team focused and helped ensure that people were making progress on their assigned tasks.

The team documented its discussions and decisions in detailed minutes. These minutes served as a record of decisions that were made over the course of the project. These proved to be valuable as new staff were added to the project they could review the past discussions to understand the basis for project decisions. The documentation provided a record that the team could use to explain to others the various alternatives that the team considered when devising the final solution.

The team wanted a strong technical basis for its decisions but at the same time understood that it was important to implement a safeguards approach as soon as possible and with limits on budgets. Also, given the high radiation levels that still exist at Unit 4, it was not feasible to initiate new studies on the conditions inside the reactor without unnecessarily incurring large radiation doses to the study teams or increasing the scope and budget of the endeavor. Consequently the team decided to perform an extensive survey of technical literature that already existed and, with the assistance of the ChNPP staff, select those studies that best characterized the true situation in Unit 4 and that had relevance for the safeguards approach. When the issue of drawing samples of the FCM, the IAEA managers pragmatically decided to use samples already taken during earlier studies stored by ChNPP and conduct an independent IAEA analysis of these samples.

The team also accepted the position that Unit 4 did not fit any of the standard facility models and therefore it was necessary to create a unique model that would fit the circumstances at ChNPP. Among the novel approaches were taking credit for the physical security system when designing the safeguards approach. This step was taken because it was impossible to perform detailed accountancy. This step also recognized the practical aspects that the IAEA did not have the time or resources to implement an entirely independent monitoring system. The team therefore considered, but ultimately did not implement, sharing radiation monitoring and video surveillance with the physical security system that was being installed at the Shelter. The IAEA expressed a desire for dual-use equipment when and where appropriate to minimize the Agency's capital investment in a safeguards monitoring system.

Through a series of meetings scheduled in 2003 and 2004, the IAEA and Ukraine exchanged technical data and discussed how best to implement safeguards that would serve the needs of the IAEA and Ukraine. For example, the team debated whether to first determine the safeguards approach or complete the DIQ. The IAEA had experience with applying safeguards when it couldn't verify the initial nuclear material declaration. This circumstance occurred at the fast breeder reactor BN350 in Kazakhstan. In this case, the IAEA measured what came into and out of the reactor and applied dual containment and surveillance to maintain continuity of knowledge. The IAEA recommended considering a similar approach for Unit 4 since they have no means to verify the material inside the Shelter. The IAEA explained that they often assisted States with developing a DIQ by providing them the IAEA's standard approach that States would subsequently use to develop their DIQ. The team understood that the circumstances at the Unit 4 were different because the standard approaches would not be applicable. If the IAEA simply asked ChNPP to submit the DIQ without further guidance, ChNPP would spend a lot of time developing a DIQ that might not meet the IAEA needs. In this case, the effort expended by the facility in developing the DIQ will be wasted, as would the IAEA's time reviewing the DIQ. The IAEA therefore developed their safeguards approach and sent ChNPP guidance on how to draft DIQ given the unique circumstances at ChNPP. This example is characteristic of the collaboration and transparency between Ukraine and the IAEA. The sides worked together to agree on a conceptual model that could be practically implemented, but developed their own detailed systems separately, thereby maintaining the IAEA's oversight role that is critical to its role in the safeguards system.

TMI-2 Nuclear Material Accountability Methodology

Nuclear material accountability for damaged reactor cores (such as TMI-2 and Fukushima) is very difficult due to the disrupted nature of the fuel and the variable composition of the debris. As such, either highly sophisticated nuclear measurement methods are required to obtain the required accuracy and precision or an alternative approach similar to that used at TMI-2 may be required. In the case of TMI-2, the Nuclear Regulatory Commission's (NRC's) regulations in Section 10 of the U.S. Code of Federal Regulations Part 70 (10 CFR 70) required that all SNM at nuclear facilities be accounted for to the nearest gram weight. The waste was destined for shipment to Idaho National Laboratory and the U.S. Department of Energy (DOE) also had similar requirements for receipt of the TMI-2 waste. Many plans were proposed for achieving this goal. However, ultimately no formal SNM accountability on a gram basis was required at the time of material shipments from TMI-2 to Idaho National Laboratory. The NRC granted the TMI-2 plant owner an exemption, wherein each shipment was covered by a DOE/NRC Form 741. The overall plan centered on having General Public Utilities Nuclear (GPUN) perform a post-defueling survey to determine the total quantity of SNM left at TMI-2 after all waste was removed. This survey was used to establish the basis for the quantity of SNM left at TMI-2. Separately, computer calculations were used to estimate the total SNM inventory in TMI-2 at the time of the accident. The difference between these two values, minus losses due to radioactive decay, was used to estimate the SNM inventory present in the reactor waste materials ultimately shipped to Idaho National Laboratory and elsewhere. This was the basis for the "one reactor core" concept. GPUN shipped "one reactor core" to Idaho National Laboratory, less small amounts of material left at the facility that could not be removed from the structure or that were trapped in filters or other process equipment as "hold-up" losses.

The primary summary of the TMI-2 nuclear accountability assessment was an internal document developed by EG&G Idaho that summarized the results of the various data sources used to assess the inventory of the TMI-2 canisters. This document "Uranium and Plutonium Content of TMI-2 Defueling Canisters"[6] is an internal Idaho National Laboratory document. This summary report provides copies of data and detailed data from General Public Utilities, including specific information from the NRC Forms 741 used to transmit the information to NRC for the shipments. Two elements are included in the summary information: (1) sampling methods used to estimate transuranic content in demineralizers and (2) other filters that generally contained small amounts of fuel and the estimates for the fuel, filter, and knockout canisters (Figure 2).

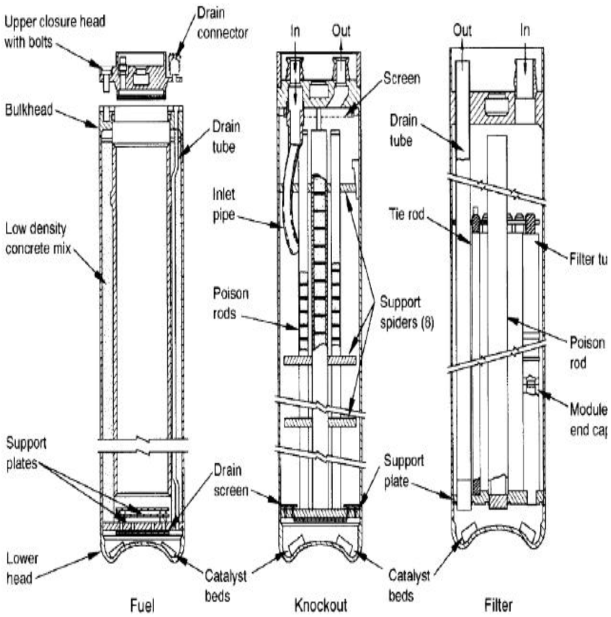

Figure 2. TMI fuel knockout and filter canisters.

The three types of canisters used to contain debris from TMI-2 were fuel canisters, knockout canisters, and filter canisters (see Figure 2). These canisters were all constructed of 304L stainless steel. The fuel canisters have a square shroud of stainless steel surrounded by a stainless steel enclosed sheet of Boral. Low-density concrete (Licon) fills the void between the shroud and the edge of the canister. The fuel canisters hold large pieces of core debris. The knockout canisters hold fine fuel particles and debris ranging in size from 140 µm up to whole fuel pellets. The filter canisters contain filters with particulates in the range of 0.5 to 800 µm. For criticality control, all canisters were filled under water in the reactor vessel, which had been flooded with water containing boron. A total of 342 TMI-2 canisters were used, which breaks down to 268 fuel canisters, 12 knockout canisters, and 62 filter canisters. Each canister was given a unique identifier as follows: fuel canisters start with D (for debris; e.g., D-301), knockout canisters start with K (e.g., K-501), and filter canisters start with F (e.g., F-401). For the purposes of the SNM canister loading analyses, canister contents were broken up into the following three classes:

1. Recognized fuel assemblies, which were known lengths of more or less intact fuel assemblies. Most were identifiable so that the original enrichment was known.
2. Structural material, which was any piece of material (for which an estimate of weight could be obtained and that contained no SNM) except surface contamination.
3. Debris that was part of any other core material in the canister. Debris included parts of fuel pins, parts of structural material, pieces of resolidified molten material, and chips from drilling or cutting operations.

The total amount of SNM fuel loaded into the canisters was based initially on the canister fill data (Table 1). These initial data indicated that the total amount of SNM in all canisters was about 2000 kg of U-235, 158.2 kg of plutonium, and 147.8 kg of combined Pu-239 and Pu-241. These initial was later modified based on the ORIGEN2 analyses by Akers and Schnitzler[7] and subsequent measurements by Akers[8] to assess the low burnup on the reactor core. Also, Akers' data on the composition of resolidified fuel material provided an estimate of the composition and fuel content of the fuel canisters. For the knockout and filter canisters, the weight, estimated fuel composition, and radionuclide content provided an estimate of transuranic content.

Extensive measurements of debris from TMI provided an assessment the SNM content of the debris including direct gamma spectrometry of cores taken from the reactor core.[9] Samples of the damaged TMI-2 reactor core were taken to spatially characterize the chemical and physical state of the degraded core. Nondestructive (e.g., visual examination, photography, sample weight, bulk sample density, and individual particle density) and destructive (e.g., optical metallography, scanning electron microscopy, and radiochemical analysis) examinations provided data on fission product release, interaction between core components, hydrogen generation, and core melt progression. The primary sources of core materials information for the reactor were sampling of the control lead screws,[10] the upper core debris bed,[11] the core bore samples from the central region of the reactor core,[12] lower head debris bed and vessel samples,[13,14,15] and examination of partial core components.[16] These sampling and analysis projects also included significant amounts of analysis through the Organization for Economic Cooperation and Development (OECD) Committee on Safety of Nuclear Installations sampling and analysis[17] and the TMI-Vessel Investigation Project (VIP) Program[13]. In addition, a report by Akers[18] summarizes the TMI characterization programs and nuclear material accountability process

Core Component	Core Material	Weight (kg)	Core Composition (%)
Fuel Assembly	Uranium	82,810	63.8
	Zirconium	23,200	17.9
	Tin	370	0.3
	Oxygen	11,300	8.7
Control Material	Silver	2,199	1.7
	Indium	412	0.3
	Cadmium	137	0.1
Structural Material	Iron	3,400	2.6
	Chromium	1,110	0.9
	Nickel	1,046	0.8
	Molybdenum	36	0.0
Miscellaneous		3,600	2.8
Total		**129,700**	**100.0**

Table 1. TMI-2 original core component composition and weight percent.

Lessons Applicable to Fukushima

There are too many differences among the ChNPP, TMI-2, and Fukushima accidents that preclude the direct application of any single approach or measurement technique developed for ChNPP or TMI-2 to Fukushima. However, there are several lessons learned that may benefit nuclear material accountancy at Fukushima.

From a ChNPP perspective, the most valuable lessons involve the carefully coordinated development of a safeguards approach, and any supporting technology, between the State and the IAEA. This helps to ensure that the approach can be implemented in a reasonable period of time while maintaining the integrity of the IAEA's role for independent safeguards verification. In the case of ChNPP safeguards approach development, the IAEA accepted at an early stage that the Unit 4 reactor core and its damaged spent fuel pools did not meet any of the standard safeguards facility models, as will be the case at Fukushima, and a unique safeguards approach needed to be created. Furthermore, the IAEA and ChNPP worked closely together to install IAEA monitoring equipment in the damaged reactor. This established a shared sense of responsibility and transparency, and ensured that both the State and IAEA had detailed knowledge of the equipment and its capabilities.

Viewed from the TMI-2 perspective, the similarities and differences between the TMI-2 and Fukushima reactors provide a basis for developing an understanding of the core degradation, handling and monitoring damaged spent fuel pools, materials behavior, core composition, and radionuclide release that can be expected when the Fukushima reactors are defueled. Further, these data provide a basis for the expected materials composition and characteristics that might be expected for the characterization of the fuel debris when it is removed and to provide a basis for simulating fuel debris characteristics for assessing the ability of SNM measurement systems.

Acknowledgements

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