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Title: ESTIMATING DISCHARGED PLUTONIUM USING MEASUREMENS OF STRUCTURAL MATERIAL ACTIVATION PRODUCTS

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Estimating Discharged Plutonium Using Measurements of Structural Material Activation Products,
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As the US and Russia move to lower numbers of deployed nuclear weapons, transparency regarding the quantity of weapons usable fissile material available in each country may become more important. In some cases detailed historical information regarding material production at individual facilities may be incomplete or not readily available, e.g., at decommissioned facilities. In such cases tools may be needed to produce estimates of aggregate material production as part of a bilateral agreement. Such measurement techniques could also provide increased confidence in declared production quantities.

The core of a nuclear reactor is usually composed of a uranium nuclear fuel and in-core structural materials. Surrounding this core is a series of external materials that are in place both to provide structural strength and stability for the core and to shield personnel from the neutron and gamma ray radiation from the reactor. These external materials are usually concrete or steel. A fluence of neutrons is imparted upon these materials proportional to the total energy generated by the reactor over the reactor's lifetime. This neutron fluence generates activation products in the structural materials. The quantity of these activation products may be a function of the activation product's characteristics (e.g., half-life), reactor power history, the dimensions of the core, the distance (and material) between the activation product location and the boundary to the reactor core, and the time since the reactor was last operated.

A three-dimensional model (Fig. 1) of the Yankee Rowe Unit 1 pressurized water reactor (PWR) core and structural materials was constructed using MCNP-4C [1]. This model was used to produce one-group neutron fluxes and cross sections for input into ORIGEN 2.1 [2]. ORIGEN was then used to simulate the plutonium production in the fuel and the activation product buildup in the structural materials (specifically the core barrel, pressure vessel, and biological shielding and structural concrete). Two simplified power histories and core loadings were assumed to explore the feasibility of this proposed technique. The first power history and core loading was similar to the actual history for the Yankee Rowe Unit 1 core (average discharge burnup of 33,000 MWd/MTU). The power history is ideal for power production use of the nuclear fuel. For this reason, this case was referred to as the "power optimized core". The second power history and core loading burns the PWR fuel to a 6% $^{240}Pu/^{239}Pu$ ratio (considered for our purposes to be weapons-grade plutonium). After the fuel reaches this burnup, the fuel is permanently discharged from the core. For this reason, this case was referred to as the "production optimized core". Both cases were simulated for a total of 4730 effective full power days.

It was found that numerous activation products would be produced in the structural materials surrounding the core. Special interest was given to the long-lived activation products that would be insignificantly affected by decay (either during fuel loadings or after permanent core shutdown). It was found that of these activation products, a small set would appear in sufficient quantity to be measurable. It was also found that a direct relationship existed between the ratio of the concentration of a long-lived activation product (e.g., ^{36}Cl) to its parent nuclide (e.g., ^{35}Cl) concentration and the total quantity of plutonium discharged by the core. This relationship for the $^{36}Cl/^{35}Cl$ ratio in the structural concrete is shown in Fig. 2. The ^{36}Cl will exist in the structural concrete at a level of 10^{12} atoms/g of concrete. These should be more than sufficient for measurement with existing mass spectroscopy instruments.

In this study, it was determined that various activation products would be produced in the structural materials surrounding a nuclear reactor in quantities which should be measurable by currently existing mass spectroscopy techniques. It was also shown that based on the quantity of these activation products measured a bounding estimate of the quantity of plutonium produced over the lifetime of the reactor can be determined.

1. J. HENDRICKS and J. BREIMEISTER, "MCNP4B2 Monte Carlo N-Particle Transport Code System," LA-12625-M, Version B, Los Alamos National Laboratory (1997).

2. A.G. CROFF, "A User's Manual for the ORIGEN2 Computer Code," ORNL/TM-7175, Oak Ridge National Laboratory (1980).

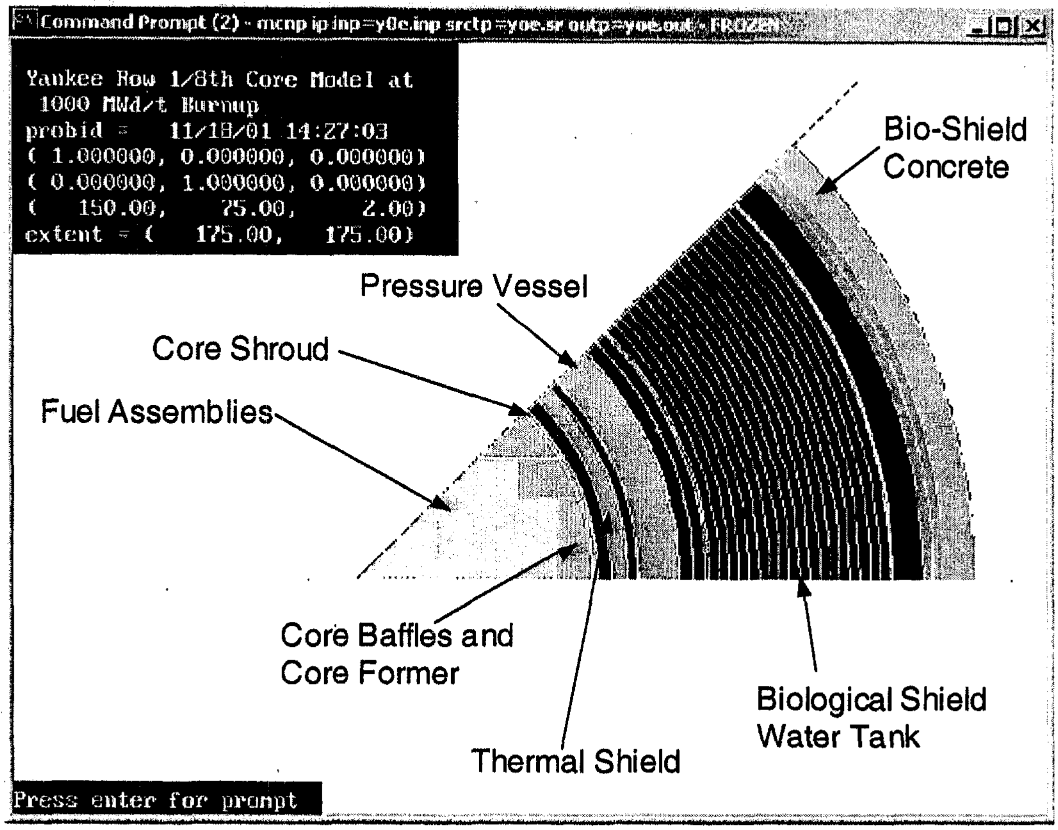

Fig. 1. Yankee Rowe Unit 1 three-dimensional MCNP-4C core model.

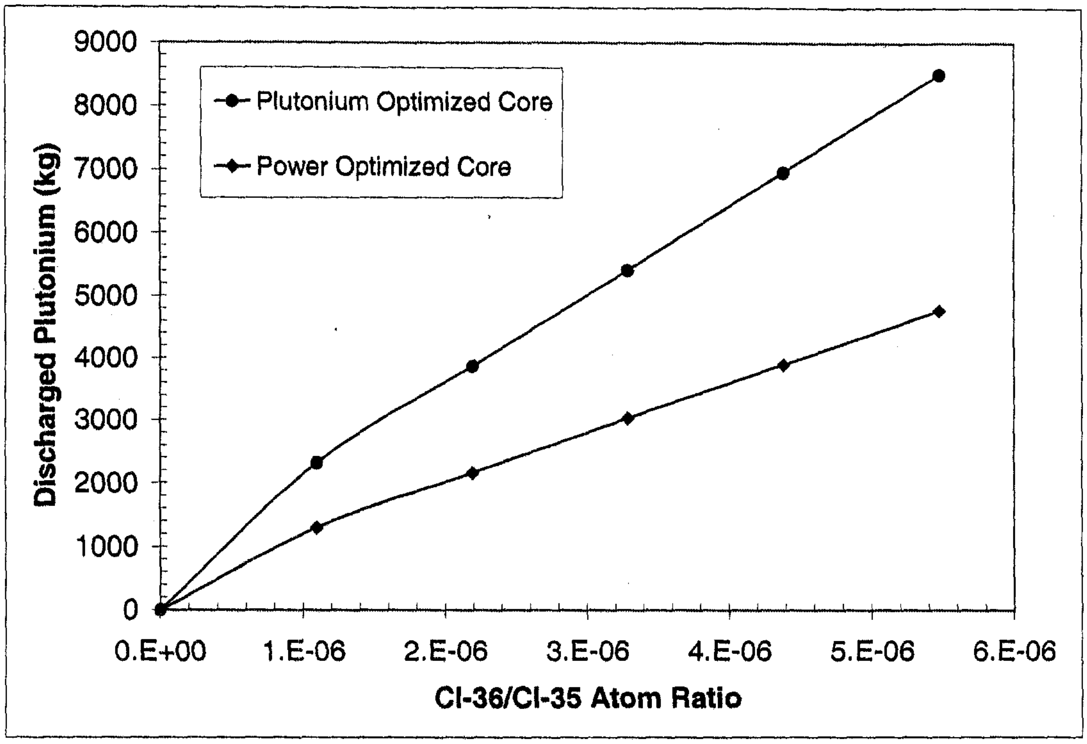

Fig. 2. Lifetime discharged plutonium estimates versus $^{36}Cl/^{35}Cl$ atom ratio for both a power optimized and plutonium optimized core.