ABSTRACT

A description is given of the ECN programme on the transmutation of the fission products technetium and iodine. Present calculations on the effect of self-shielding, experimental studies for sample characterization and sample selection and the design of an irradiation facility for use in the High Flux Reactor (Petten) are discussed.

INTRODUCTION

Partitioning and transmutation (P&T) of actinides and fission products are attracting considerable attention at present as an option to reduce the long-term radiological hazard of the high-level nuclear waste (HLW).1,2 The prospects of P&T are the subject of worldwide studies3-7 and there is general agreement that the implementation of P&T in waste management is technically feasible. However, much research needs still to be done to improve the partitioning (reprocessing) of the radionuclides intended for P&T as well as to resolve the technological problems of the transmutation, to make the process industrially attractive.

The beta-emitting fission products technetium (99Tc, half-life 2.13 × 107 year) and iodine (129I, half-life 1.57 × 107 year) are among the important long-lived nuclides in high-level waste, that dominate the beta radiotoxicity for more than a million years. Although the beta toxicity will remain far below the alpha toxicity of the actinides, the risk during-storage, defined as the product of toxicity and chemical mobility, will be relatively high because technetium (in the form of the TcO4− ion) and iodine are rather mobile in geochemical environments of underground waste repositories.2,8

Transmutation of 99Tc and 129I by neutron capture as a result of irradiation in nuclear reactors will yield the stable isotopes 100Ru and 130Xe, respectively. However, due to the small neutron cross-sections, the transmutation efficiency in LWRs is low: only a few percent of the technetium is transformed in a year. For iodine the efficiency is only little better.9 High-flux thermal reactors such as the CANDU or moderated assemblies in fast power reactors are therefore more appropriate devices for the transmutation of these fission products. Also intense neutron fields produced by future accelerator-based thermal reactor systems are of interest.10 Due to the low transmutation efficiencies, multiple reprocessing of the irradiated inventory will be required in practical cases. This makes the transmutation option with technetium homogeneously distributed through the fresh fuel not attractive since considerable effort is required to separate this element from spent fuel. Irradiation in special targets (heterogeneous transmutation) is therefore the most likely technique to be applied for both fission products.

In 1991 ECN has started a research programme on the transmutation of technetium and iodine which will be described here. In this study the physical, chemical and technological aspects of the heterogeneous transmutation in existing or future-generation power reactors are being considered, with special emphasis on the determination of transmutation efficiencies to evaluate calculational methods and on the study of the material behaviour. A facility for irradiation experiments in the High Flux Reactor (HFR Petten) is being constructed and the first experiments on milligram-sized samples are planned in 1994. To our knowledge few of such experiments have been performed. Wootan et al.11 have described transmutation experiments on milligram-sized samples in a Fast Flux Test Facility which yielded calculation-to-experiment transmutation ratios of 0.80 for 99Tc and 0.86 for 129I. These experiments were, however, not primarily aimed at the study of materials asp ects.

TECHNETIUM

Chemical aspects

Technetium is recovered from the PUREX waste stream as a salt, generally ammonium pertechnetate (NH4TcO4).12 This material must be converted to a suitable target material before...