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# Beratungsunterlage zu TOP 3 der 6. Sitzung

## Zusammenfassung des Kurzvortrags von Prof. Dr. Reto Gieré

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# Nuclear Waste Forms and Natural Analogs

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This presentation will discuss research results and recommendations, which are based on a long-term collaboration between scientists at the University of Freiburg, the Australian Nuclear Science and Technology Organisation (ANSTO) in Menai (New South Wales, Australia), the Natural History Museum in London (UK), the Pacific Northwest National Laboratory (PNNL) in Richland (Washington, USA), and Purdue University in West Lafayette (Indiana, USA).

The main thesis behind natural analog studies is the very fact that uranium ore bodies and uranium-rich minerals have survived intact for eons, despite significant changes in climate and geological conditions. Natural analogs contain information that can be of use in the design and construction of anthropogenic systems, particularly high-level waste (HLW) repositories.

The unique role of natural analog systems is to provide a link between the very short time scales of field and laboratory studies and the enormous geological periods over which the results of such studies are extrapolated in performance assessments (PA) of an HLW repository. Laboratory experiments, for example, provide data on elemental release rates under controlled conditions, but often fail to elucidate the alteration mechanisms, even when reaction products are easily observed. Studies of minerals from various geological environments, on the other hand, may reveal quantitative data on the geochemical alteration mechanisms, but only limited inferences can be made concerning the reaction kinetics.

Further important roles of natural analogs are their ability to give scientists access to the true complexity and heterogeneity of real systems and the ease with which explanation by analogy is generally understood compared to the subtleties of mathematical PA models.

Natural analogs are often presented as key components of national HLW disposal programs. One of the most extreme cases is Japan, where this requirement is explicitly stated in the nuclear law for the disposal of special wastes (see Alexander *et al*., 2006).

The purpose of this short presentation is to provide an overview of different waste-form types for HLW (for details, see Stefanovsky *et al*., 2004) and their geochemical behavior in the geological environment (for details, see Lumpkin *et al*., 2004, 2014).

**Waste Forms**

In addition to the direct disposal of spent nuclear fuel, borosilicate glass and polyphase or single-phase ceramic waste forms have been considered as options for the disposal of nuclear waste in geological repositories. Glasses, ceramics, and glass-ceramics are suitable waste forms for HLW, whereas cement, bitumen, glass, glassy slags, and ceramics are suitable for low- and intermediate-level waste (LILW).

Ceramic waste forms have the highest chemical durability and radiation resistance, and thus are recommended for HLW and actinide immobilization. Most radiation-resistant are materials based on phases with a fluorite-related structure (*e.g.*, cubic zirconia-based solid solutions, pyrochlore, zirconolite,). Glass is also a suitable matrix for HLW that contains fission and corrosion products, as well as process contaminants such as Na salts.

Within the framework of the HLW partitioning concept providing separation of short-lived (Cs, Sr) and long-lived (rare earth element – actinide) fractions, glass may be used for immobilization of the Cs-Sr-bearing fraction, whereas the rare earth – actinide fraction may be best incorporated into ceramics (*e.g*., titanate ceramics).

**Geochemical Behavior**

In the case of titanate ceramics, we are primarily concerned with the oxide phases brannerite, hollandite, perovskite, pyrochlore, and zirconolite, but also with other phases, such as, apatite, zircon, monazite, and zirconate defect-fluorite/pyrochlore structure types. Of these structure types, natural samples exist for all but the zirconate defect-fluorite/pyrochlore phases.

For monazite, zircon, and zirconolite, existing data demonstrate acceptable laboratory leach rates together with limited geochemical alteration in natural environments.

Hollandite appears to be a highly durable host phase for radioactive Cs, but investigations of natural samples are needed in order to study the long-term behavior.

Pyrochlores, both titanate and zirconate, also have acceptable leach rates, but long-term data are only available for titanate pyrochlores (together with Nb and Ta rich varieties).

Spent fuel can be regarded as a special HWL ceramic, and for this type, nature provides excellent examples, which demonstrate clays are suitable are as host-rocks for geological time scales.

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