Innovative Conditioning Procedures for the Generation of Radioactive Waste Products which are Stable for Intermediate Storage or Repository-Independent in Final Storage

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ABSTRACT

The German Federal Government aims at a future final storage site for all kinds of radioactive waste within 30 years. Existing and newly-produced radioactive waste therefore has to be stored in interim storage facilities over very long periods of time.

At present, most German radioactive waste or waste packages are produced and qualified according to the acceptance criteria of the projected final repository KONRAD.[1] Nevertheless, conditioning strategies for crude radioactive waste have to take into account the open question of the future repository site as well as requirements for long-term interim storage.

The Quality Control Group for Radioactive Waste (in German: Produktkontrollstelle fuer radioaktive Abfaelle - PKS) works as an independent expert organisation for the quality checking of radioactive waste packages as well as evaluating conditioning procedures for waste containers suitable for final storage on behalf of the Federal Office for Radiation Protection (in German: Bundesamt fuer Strahlenschutz – BfS). The Institute for Safety Research and Reactor Technology (in German: Institut fuer Sicherheitsforschung und Reaktortechnik – ISR) of the Research Centre Juelich investigates scientific/technical problems of nuclear disposal, especially in the field of waste treatment. In this context, ISR and PKS investigated and/or evaluated innovative procedures, by means of which radioactive waste flows may be minimized and rendered inert. QSA Global (formerly: AEA Technology QSA) conditions radioactive waste of German users from the fields of medicine, research and industry as well as from its own radioactive source production and operates an intermediate storage facility for radioactive waste containers.

This poster deals with the characteristics and possible applications of new waste fixation media on the basis of organic and inorganic mineral polymers; with the approach of producing inherently safe waste forms for various geological formations. Plasma technology and inorganic additives produce volume reduced glasses.

Organic polymers evaluated are polysiloxane compounds with additives like barium sulfate, lead dioxide or others, depending on the specific requirements. As a counterpart to organic polymers, mineral polymers are based on silica and alumina, exhibiting better mechanical and thermal properties, as well as higher durability, compared with concrete. Thus QSA Global uses mineral polymers for packing radioactive waste containers, if high safety requirements have to be fulfilled like the waste acceptance

criteria for the KONRAD repository. (Plasma products so far generated in experiments resemble natural obsidian, a highly inert and stable volcanic glass.)

INTRODUCTION

The German Federal Government aims at establishing a single future repository in a geological formation still to be defined, into which all types of radioactive waste can be stored. Until its construction and commissioning, existing and newly arising radioactive waste must be interim-stored over probably very long periods of time. This does not only concern the operational and, as a rule, homogeneous waste from nuclear power plants, but also the heterogeneous waste streams from medicine, research and industry and from the dismantling of nuclear installations and facilities. They account for the by far largest volume consisting of "non-heat-generating radioactive waste" and "radioactive residues".

Conventional conditioning techniques convert the above crude waste into waste products, which may pose problems in long-term interim storage and with respect to later final disposal capability in a still undefined geological formation. For this reason, new and complementary conditioning techniques are needed. They should convert waste streams critical in interim or final storage into chemically inert and repository-independent waste products with long-term stability. At the same time they should not interfere with acceptance criteria for a future repository not yet precisely defined today.

UTILIZATION OF GEOPOLYMERS FOR WASTE-PACKING AT QSA GLOBAL

The former iron ore mine KONRAD, located near the town of Salzgitter in the state of Lower Saxony in Germany, is projected to be the future German final repository for low and intermediate level radwaste and radwaste products. For the final storage in KONRAD, basically specific containers are intended as waste packages. If the nuclide inventory exceeds 10 percent of the limits for acceptance at KONRAD, containers have to be carried out in a specific (safe) form, stable against mechanical damage even after crashes and without hazardous release of radioactivity in case of a burning underground container transporter.

So far, safe KONRAD containers have been developed and approved only for cemented radwaste coming from nuclear power plants and containing uranium and/or plutonium. In cooperation with Eisenwerke Bassum GmbH as an important German manufacturer, QSA Global has developed a concept how a KONRAD container can be loaded with all kinds of activity originating from medicine, research and industry in Germany.

For the safe fixation or cementation of radioactive waste, a specific concrete on the basis of alumino silicate is used, named Geopolymere[R].[2] This concrete is already used for the immobilization of harmful substances, pollutants and naturally occurring radioactive material (NORM).

Showing thixotropic properties Geopolymers[R] is characterized by very favourable advantages in radwaste conditioning. Thixotropy is defined as the property of a non-newtonian pseudoplastic fluid to show a time-dependent change in viscosity with the applied shear-force. Thus already plastic to stiff geopolymers mixtures will become fluid again after the input of mechanical energy, for example by shaking or vibration, and they will remain in this state during a significant relaxation time. In practice hollow spaces of radioactively contaminated components can be much better filled with Geopolymers[R] than it is possible with customary concrete. In addition, radioactive waste packages can be filled with Geopolymers[R] via pipes with small diameter. Bubbles are transported to the concrete surface during processing thus resulting in a very homogeneous concrete matrix.

Fig. 1 Containers Type IV, licenced for final storage in the projected repository KONRAD

Concerning long-term interim storage Geopolymers^{R} exhibit high physical and chemical stability surpassing usual concrete, for instance against corrosion by sulphate, a common concrete poison. Cured Geopolymers^{R} when they become wet, absorb water in a micro-cellular structure with high capillary pressure. Thus, even at very low temperatures, a geopolymeric matrix will not disintegrate due to freezing water inside the waste product.

Basic material for Geopolymers^{R} is potassium alumino silicate. Mixed with aqueous potassium silicate (soluble glas) in alkaline media potassium alumino silicate binds off forming inorganic polymeric structures similar to zeolites. Hence hardened Geopolymers^{R} is a condensation product of only inorganic compounds without organic parts. As for the chemical structure, Geopolymers^{R} can therefore be compared with ceramics, but with the advantage of being produced at normal temperatures using standard techniques of concrete handling.

For containment the approved KONRAD container type IV of the Eisenwerke Bassum GmbH is used. Side, top and bottom walls of the container are lined for thermal protection with Geopolymers^{R} or concrete layers. It contains an inner box (inliner) which can be loaded with waste packages or waste compounds both filled with and fixed in Geopolymers^{R}. As waste packages, standard waste drums or box-shaped packages are intended. By stacking the box-shaped packages in the container the packing density of the radioactive waste can be increased, minimizing the volume and thus the costs for final storage.

On behalf of the German Federal Office for Radiation Protection (BfS), the container described was examined by both the German Federal Office for Material Science and Testing (BAM) in Berlin and the Quality Control Group for Radioactive Waste (PKS) in Juelich [3]resulting in recommendations to license it for final storage in KONRAD. The possible release of radioactivity during a fire and cooling scenario was modelled and calculated by the Institute for Safety Technology GmbH (ISTec) in Cologne.

HIGH-TEMPERATURE PLASMA TECHNOLOGIES FOR WASTE TREATMENT

Present treatment techniques for crude waste generally involve machining and fixation in a cement matrix, sometimes in combination with preceding combustion and/or subsequent drying. However, this does not decisively suppress chemical reactivity. It may even be increased. This applies, in particular, to old waste, most of which is mixed waste and whose packaging frequently shows internal corrosion and outgassing. Such waste can be transformed by thermal decomposition in a plasma arc, simultaneously reducing its volume, into long-term stable, near-natural glassy products. They can be emplaced in many host rocks in a retrievable and package-independent form. The products thus resemble natural obsidian, an inert and geochemically stable extrusive rock. Last but not least, plasma techniques provide a means of selectively confecting waste products according to storage requirements by varying the process parameters.

Plasma technologies have already been successfully applied in the treatment of hazardous and toxic waste. In the conventional field, there are already various techniques worldwide that could be or are suitable for adaptation to the conditions of operation with radioactive waste. In the USA, the process-engineering development of plasma machines for the treatment of mixed waste has left the field of eligible research and is now commercially pursued. France recognizes a very high potential for the plasma-thermal treatment of radioactive waste and pursues relevant scientific and technical investigations at the CEA research centre, Cadarache. In Switzerland, a prototype plasma machine for the conditioning of radioactive waste is in routine operation in the national ZWILAG interim storage facility at Wuerenlingen. In Taiwan, a facility for the treatment of radioactive waste drums is already in operation without any known problems.

On the whole, an international innovation trend can be recognized, which emphasizes the advantages of plasma products with a view to product stability, considerably reduced storage volume and large repository independence. The new technique and its development and application is characterized by great dynamics worldwide. In the Federal Republic of Germany, ISR is the only research institute concerned with the applications of plasma technologies for nuclear waste management.

Against the international background, it therefore appears meaningful to investigate the possibilities of plasma technologies for German waste under realistic conditions in order to create the fundamentals and know-how for possible application. The aim research work at ISR was to determine and optimize the properties of plasma products and of the process-engineering parameters.

In first experiments, performed in cooperation with RWTH Aachen University of Technology, the thermal treatment of non-combustible simulates of nuclear power plant waste was examined at the RWTH's plasma unit. This was done in close cooperation with the Obrigheim nuclear power station. Fig. 2 shows typical products from plasma-thermal waste treatment, whose properties are comparable to obsidian occurring in nature.

Fig. 2: Typical products from plasma-thermal treatment

APPLICATION OF POLYSILOXANES IN NUCLEAR INDUSTRY

An important problem of long-term storage is the corrosion of storage containers. The presence of water especially during cementation significantly accelerates the container corrosion. Moreover, the handling and transport can lead to damaging of container walls and facilitate the metal corrosion. Often, existing containers show significant corrosion damages already after few years. Therefore a need of advanced protective coatings for the storage containers exists.

Polysiloxane show good physical and chemical properties and seem to be an adequate material for treatment of radioactive wastes. It can be used as an outer or inner coating for the purpose of mechanical and corrosion protection. Investigations at ISR were carried out using different polysiloxane types and supplied the results with regard to corrosion protection, mechanical properties, water vapour diffusion, radiation resistance and application techniques.

One of the advantages of this material is the possibility to form thick layers (e.g. 5 mm) which supply a protection against mechanical damage. Due to its chemical structure, this material is hydrophobic and waterproof. The water vapour diffusion was investigated and is higher than in other elastomer materials. Zinc powder and micaceous iron oxide were therefore successfully used as rust-protecting additives in different polysiloxanes.

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