CERAMICS AS POSSIBLE FUTURE ALTERNATIVE MATERIALS FOR THE HLW OVERPACK (CIGEO PROJECT)

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Context and objectives

Why Ceramic solution for HLW overpack?
Cigeo Project: the French deep geological disposal
A general view at the end of operations

2 surface facilities:
- Receiving, inspecting, and preparing packages
- Shafts for construction work and "conventional" installations

- If licensing, Cigéo will be located at the edge of Meuse and Haute Marne districts in eastern France
Cigeo Project
Major Milestones

ACT ON THE TERMS OF CONSTRUCTION OF CIGEO AND ITS ACCRUAL

2016

SUBMISSION OF
CONSTRUCTION LICENCE APPLICATION

2019

CONSTRUCTION LICENCE GRANTED BY CIGEO

2022

INDUSTRIAL OPERATIONS LICENSE

2030

INDUSTRIAL OPERATIONS LICENSE

2035

ACT AUTHORIZING DEFINITIVE CLOSURE OF THE REPOSITORY

2150

DESIGN

ASSESSMENT

INITIAL CONSTRUCTION

INDUSTRIAL PILOT PHASE

PROGRESSIVE CONSTRUCTION & OPERATION

CLOSURE & MONITORING

SITE CHARACTERISATION, RESCUE ARCHAEOLOGY & PRELIMINARY WORK

(Site survey, data collection, preparation of project and power connections, etc.)

SUBMISSION OF REPORTS

- Safety plans
- Technical reliability options
- Proposed master plan for facility operation

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Radewaste for Cigeo Project
High Level Waste (HLW) and Intermediate Level Long Life Waste (IL-LLW)

1- Waste from spent fuel treatment (HLW and IL-LLW)

Forecast volumes: about 72,000 m³ for IL-LLW (of which 60% already produced) and 10,000 m³ for HLW (of which 30% already produced)

Spent Fuel disposal is studied in adaptability of Cigeo Project
Cigeo project
Reference solution of HLW disposal cell for licensing application

Primary package conditioning in carbon steel over-pack
  o To prevent water contact with glass during 500 y in post closure
    • Low carbon steel; thickness ~55 mm

Horizontal disposal cell with carbon steel sleeve
  o To emplace et remove HLW disposal packages during operating period
    • Length: 100 à 150 m; diameter: 70 cm
Cigeo project
A progressive and incremental development...

- Installation for licensing application, and HA0 pilote (years 2030)
  HA0 pilote zone and 4 IL-LLW disposal cells

- A secular exploitation
- A progressive construction

Installation at the end of operations (years 2140)
stockage de l’inventaire de référence

HLW 1/2 zone
2080 ➔ 2140

IL-LLW zone ➔ 2080
Cigeo project
A progressive and incremental development...

The reference concept for licensing application, and HA0 pilot zone

The possibility of concept evolution, in link with time schedule of HLW1/2 disposal operating

Forward-looking solution for the overpack material: Ceramics
  - Inert materials
    - Preservation of a neutral environment for the other components glass and clay
  - No H\textsubscript{2} production
  - R&D Program since 2007
R&D Development

Ceramic Overpack design & manufacturing

- Feasibility of large shaped watertight ceramic pieces?
- Solution for joining these pieces at moderate temperature (600-700°C)?
- Suitable mechanical strength?
Ceramic overpack design

1- Development of a watertight alumina-silica ceramic
   • Elaboration of 5 ceramic grades on alumina-silica diagram
     • With a range of alumina content (60 to 80 %)
     • With test of quartz influence
   
   Composite effect of mullite needles ?

   • Issues to consider for ceramic selection
     • Suitable mechanical strength 100 < flexion failure < 140 Mpa
     • Suitable chemical durability
     • Reproducibility of the material properties
     • Open porosities ≤0.3 %

A ceramic in the intermediate range of alumina content (72%) has been selected.
Ceramic overpack design

Mechanical characterization of the selected ceramic

- Application of Weibull methodology (dispersion)
  - 4 points bending tensile strength tests
  - Weibull modulus: $m = 15$
    (Modulus scales from 5 for brick, up to 100 for steel)

- Other mechanical data acquired
  - Young modulus: 146 Gpa
  - Shearing modulus: 61 Gpa
  - Poisson coefficient: 0.19
  - Tenacity: $K_{IC}$ within 1 to 3 MPa m$^{1/2}$

**Good W.mod. for a brittle material**
Ceramic overpack design

2-Design adapted to the mechanical properties of a ceramic

- Loading cases
  - Water pressure → Isotropic load
  - Lithostatic load → Anisotropic load

- Shape: hemi-spherical extremities

- Canister retrieval assumed without traction strength (not defined yet)

- Allowable strength taking into account:
  - failure ratio of 1/21000 – Dossier 2005 basis
  - Loading case
  - Volume and shape of the overpack
  - Weibull modulus

- Finite element calculations

A thickness value of 40 mm complies with requirements for an isotropic load (12 or 16.3 MPa)
Ceramic overpack design

3 - Adjustment of slipcasting and sintering parameters to realize a thick ceramic part (40 mm)

4 - Realization of an ½ scale overpack
   o Length : 70 cm
   o Thickness : 4 cm

Next step : sealing process
R&D Development

Sealing Process
Ceramic overpack sealing challenge

Statement of work

- Sealing zone must have the same properties as the body
  - durability
  - mechanical strength
  - gas-tightness

- Preservation of the nuclear glass matrix \((T_{\text{max}}<450^\circ\text{C})\)
  - Heating only on the sealing zone
  - Temperature limited at \(700-800^\circ\text{C}\) at the extrado of the ceramic body

Priority Action since 2010: development of suitable sealing (process and design) at low temperature for ceramic pieces
Ceramic overpack sealing R&D

Since 2010, R&D studies focused on joining process

- **Identification of a bonding technology (expert group):**
  - Solid state joining techniques as diffusion bonding appear highly favourable
  - Materials close to glassy phase has to be used as a glass interlayer

- **Necessary optimization:** find an appropriate combination of pressure, heat and time
  - Laser heating: probably not suited to the cylinder thickness
  - Induction heating: the more common heating technique but need the use of a susceptor because of the poor electric susceptibility of alumina ceramic
    - Joining of 10 mm thick pieces at 1000°C
    - Steel or graphite susceptors not suitable: necessary to test another material
    - What about thicker pieces (40-50 mm)?
  - Microwave heating: most likely to be scalable to the size of the overpack
Microwave heating for ceramic overpack sealing: Feasibility approach

Identification of compliant glassy phases as joining material

- Melting temperature: 500-1000°C
- Coefficient of Thermal Expansion compliant with Andra Ceramic one (CTE = 61 10^{-7} K^{-1} 25-200°C): 55-65 10^{-7} K^{-1}

3 compositions tested

<table>
<thead>
<tr>
<th>Supplier</th>
<th>Ref.</th>
<th>SiO2 (%m)</th>
<th>B2O3 (%m)</th>
<th>Al2O3 (%m)</th>
<th>ZnO (%m)</th>
<th>CaO (%m)</th>
<th>Autres (%m)</th>
<th>CTE (25-300°C) (*10^{-7} K^{-1})</th>
<th>Melting point (°C)</th>
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<tbody>
<tr>
<td>Johnson Matthey</td>
<td>1*</td>
<td>2*</td>
<td>3*</td>
<td></td>
<td></td>
<td></td>
<td>BaO 4*</td>
<td>62</td>
<td>920</td>
</tr>
<tr>
<td>Emaux Soyer</td>
<td>REL063</td>
<td>1*</td>
<td>2*</td>
<td>3*</td>
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<td>660</td>
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<tr>
<td>Emaux Soyer</td>
<td>REL064</td>
<td>3*</td>
<td>1*</td>
<td></td>
<td></td>
<td></td>
<td>Bi2O3 2*</td>
<td>61</td>
<td>520</td>
</tr>
</tbody>
</table>

* 1 à 4: sorting in descending order
Microwave heating for ceramic overpack sealing: Feasibility approach

First tests of microwave heating at laboratory scale

- Interaction between ceramic and microwave? Use of adding susceptor?
- Interaction between sealing glasses and microwave?
Microwave heating for ceramic overpack sealing: Some results

Direct microwave heating of ceramics

All glasses interact with microwave: REL63 < JM << REL64
Microwave heating for ceramic overpack sealing: Some results

Feasibility of assembling by microwave heating achieved

Defaults (bubbles) observed in the glassy phases
  - Curing of the enamel induces degassing
  - Bubbles are trapped in the assembly

How can be optimized the properties of the assembly and the process?

Development of a two step heating process
  - Enameling of the ceramic sealing surfaces (industrial process)
    - Spraying of the glass slurry on ceramic
    - First heating of the enamel in a conventional furnace
  - Assembling of the enameled pieces by microwave heating

Joining of enameled ceramics demonstrated by microwave heating
Better microstructure of the sealing material
Microwave heating for ceramic overpack sealing: Some results: mechanical properties

Samples 4x3x20 mm³
4 point bending tests

Breaking stress (MPa)

**Pencil brushed glassy slurry**

<table>
<thead>
<tr>
<th></th>
<th>REL 63 GLASS</th>
<th>REL 64 GLASS</th>
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<tbody>
<tr>
<td>Direct microwave joining</td>
<td>15,7 ± 0,6</td>
<td>26,7 ± 5,2</td>
</tr>
<tr>
<td>Two step process</td>
<td>21,5 ± 2,0</td>
<td>76 ± 17</td>
</tr>
</tbody>
</table>

**Sprayed glassy slurry**

<table>
<thead>
<tr>
<th></th>
<th>REL 63 GLASS</th>
<th>REL 64 GLASS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Two step process</td>
<td>147 ± 26</td>
<td>96 ± 34</td>
</tr>
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</table>

Ceramic: 135 ± 4MPa

Improvement of the sealing quality with the new process
Microwave heating for ceramic overpack sealing: Next step…

Is it possible to join ceramics pieces with an external microwave heating? (TRL 3 to TRL 4)
  o Consulting of an industrial microwave furnace manufacturer (SAIREM)
  o Development of a concept of microwave furnace for ceramic overpack sealing

Changing the scale and forms of the sample
  o Ring forms
  o ¼ and ½ scale

Development of a microwave furnace prototype to test ring assembling

Study of new sealing materials: ceramic-glass
  o Better mechanical and lixiviation properties
  o Question: how does this kind of materials react with microwave?
SCELLMA Project: SEALING ceramic overpacks by plasma torch

Aim: to develop a sealing process for the ceramic disposal containers using thermal plasma technology

4 Partners, duration: 46 months

Working axes:
- Sealing area design for plasma technology
- Identification of materials (ceramic, glass) for the sealing
  - Chemical and process compatibility
  - Lixiviation performances
- Adaptation of projection parameters on laboratory pieces
  - Gas, temperature
  - Projection speed
- Sealing of a demonstrator
Conclusions

Overpack design
 o Further work required for the sizing of the overpack with more accurate and updated mechanical calculations (anisotropic load in progress)
 o Ceramic composition and implementation optimization for a full size demonstrator
 o Retriavibility of ceramic overpack development

Sealing process
 o Microwave solution:
   • Feasibility at lab scale (TRL 2-3)
   • To continue : testing local heating in a prototype of microwave furnace
 o Optimization of sealing materials : use of ceramic-glass?
 o Plasma torch technology : feasibility to proof ( thermomechanical )
Thank you for your attention