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

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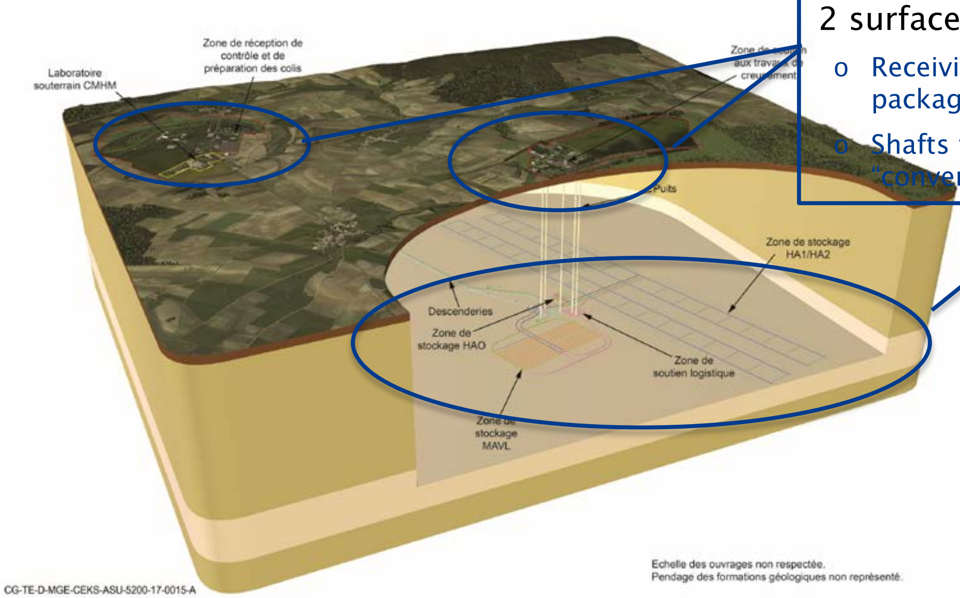
# Context and objectives

## Why Ceramic solution for HLW overpack?

# Cigeo Project : the French deep geological disposal

## A general view at the end of operations

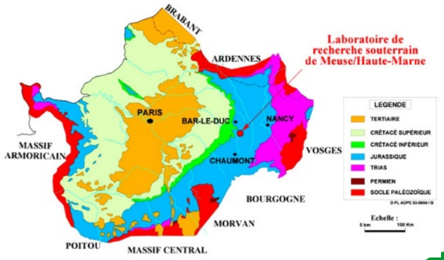



2 surface facilities:

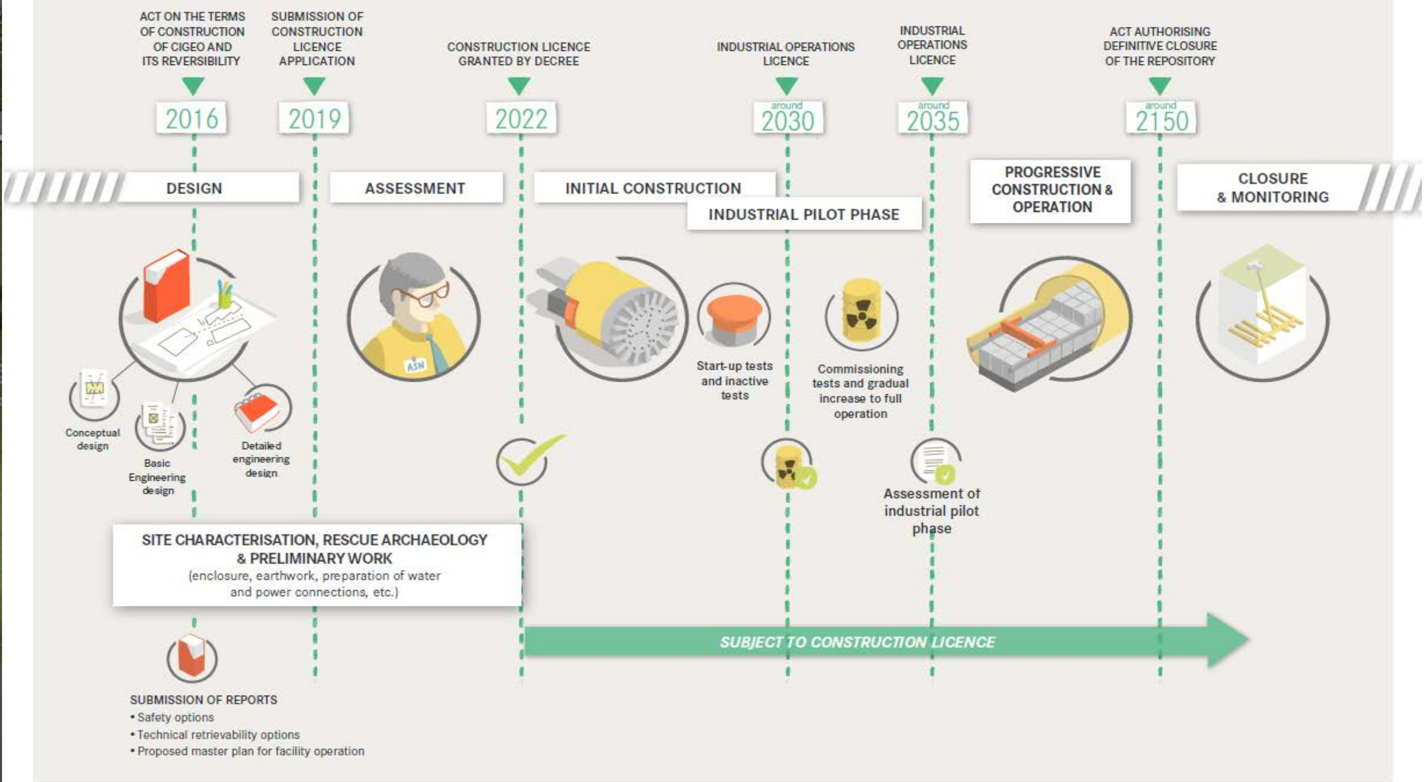
- Receiving, inspecting, and preparing packages
- Shafts for construction work and "conventional" installations

Underground facility in clay (500m depth)

- If licensing, Cigéo will be located a the edge of Meuse and Haute Marne districts in eastern France



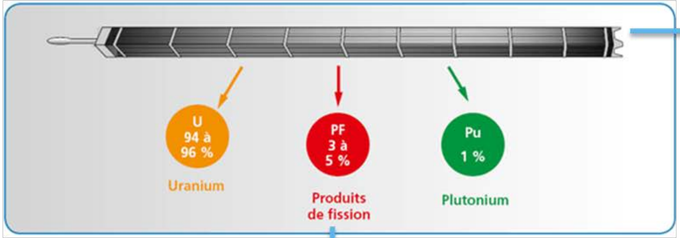
# Cigeo Project
## Major Milestones



# 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)




Waste metal from the structures surrounding spent fuel

Metallic package for vitrified HLW

### 2- Technology waste, research activities and legacy waste (IL-LLW)

**Forecast volumes :** about 72, 000 m$^3$ for **IL-LLW** (of which 60% already produced) et 10, 000 m$^3$ for **HLW** (of which 30% already produced)
Spent Fuel disposal is studied in adaptability of Cigeo Project





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# Cigeo project

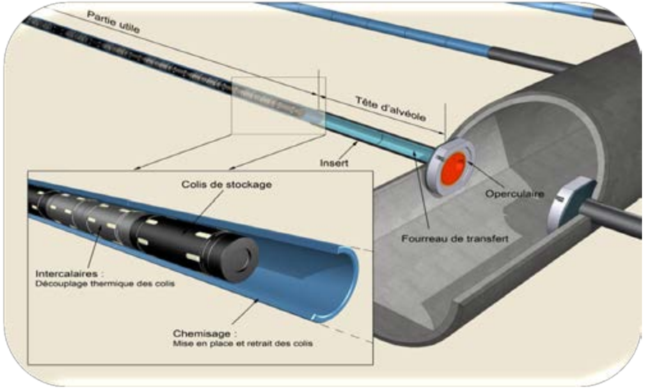
## Reference solution of HLW disposal cell for licensing application

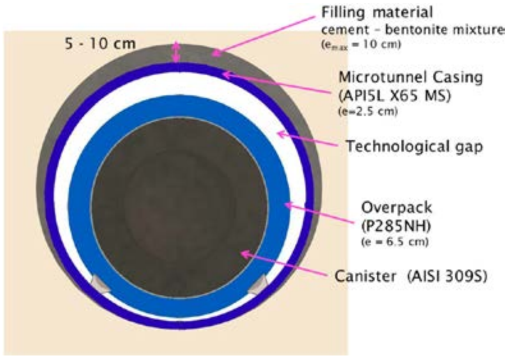
Primary package conditioning in carbon steel over-pack

- To prevent water contact with glass during 500 y in post closure
  - Low carbon steel ; thickness ~55 mm

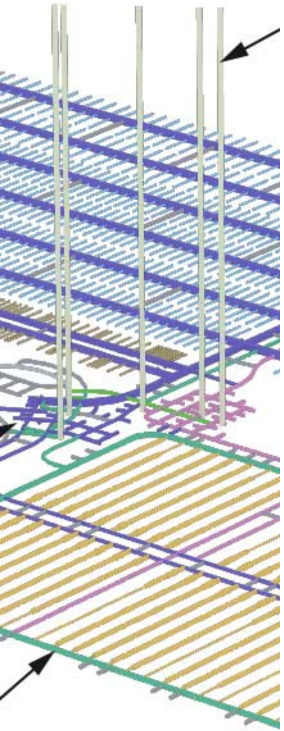
Horizontal disposal cell with carbon steel sleeve

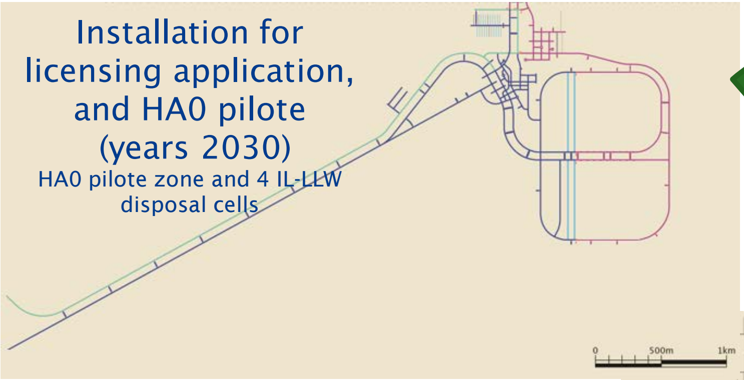
- To emplace et remove HLW disposal packages during operating period
  - Length: 100 à 150 m; diameter: 70 cm

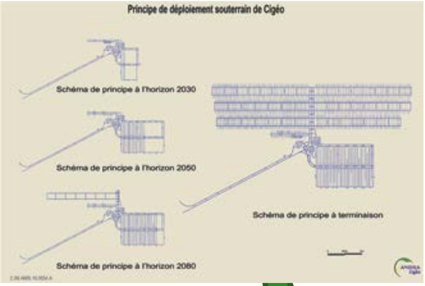



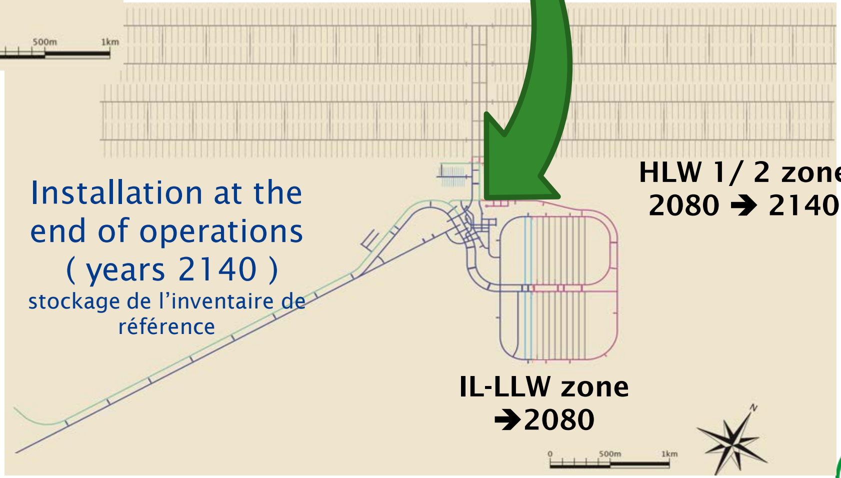
# Cigeo project
## A progressive and incremental development…



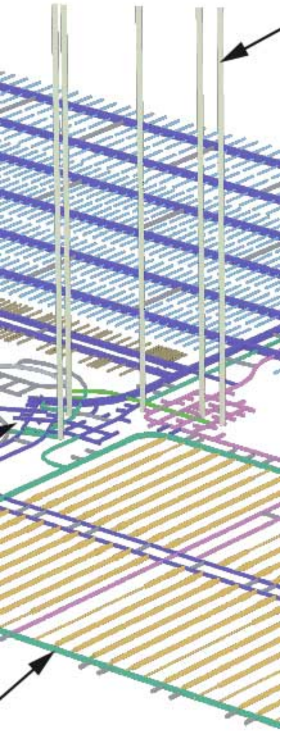



- A secular exploitation
- A progressive construction



# 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 $H_2$ production
- R&D Program since 2007





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# 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

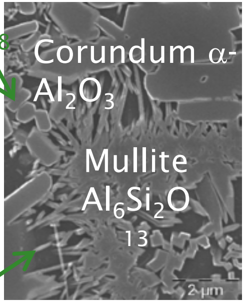
## 1- Development of a watertight alumina-silica ceramic

- o 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 ?***

- o 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

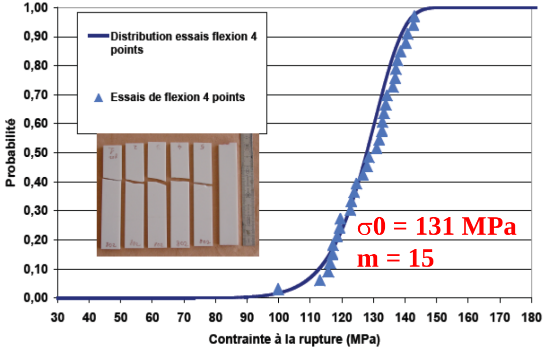
## Mechanical characterization of the selected ceramic

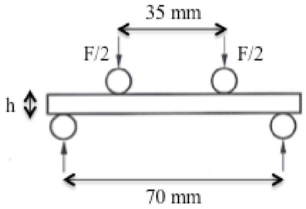
- Application of Weibull methodology (dispersion)
  - 4 points bending tensile strength tests
  - Weibull modulus: m = 15

    (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 : $K_{1C}$ within 1 to 3 MPa $m^{1/2}$

**Good W.mod. for a brittle material**

Weibull modulus (30 samples)





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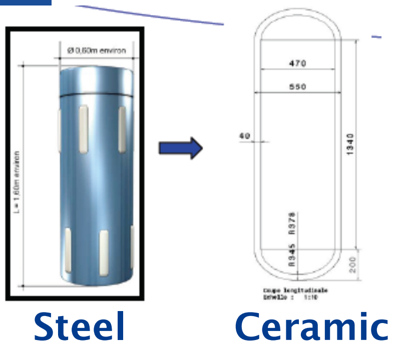
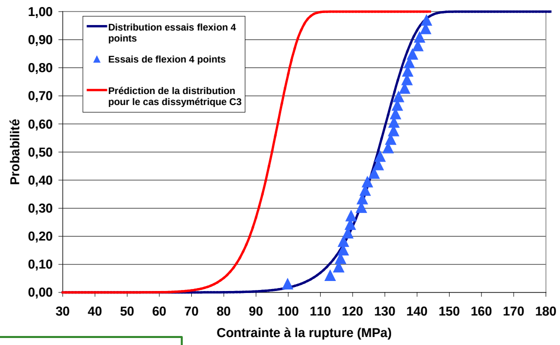
# Ceramic overpack design

2-Design adapted to the mechanical properties of a ceramic

- o Loading cases
  - Water pressure → Isotropic load
  - Lithostatic load → Anisotropic load
- o Shape : hemi-spherical extremities
- o Canister retrieval assumed without traction strength (not defined yet)
- o Allowable strength taking into account :
  - failure ratio of 1/21000 – Dossier 2005 basis
  - Loading case
  - Volume and shape of the overpack
  - Weibull modulus
- o 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

- Length : 70 cm
- Thickness : 4 cm


**Next step : sealing process**

# R&D Development

## Sealing Process





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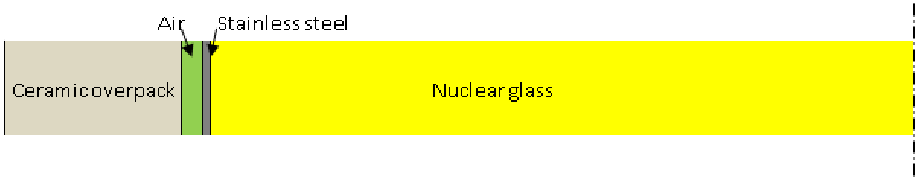
# Ceramic overpack sealing challenge

Statement of work

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

- o Preservation of the nuclear glass matrix (Tmax<450°C)
  - Heating only on the sealing zone
  - Temperature limited at 700-800)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





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# 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 $10^{-7}$ $K^{-1}$

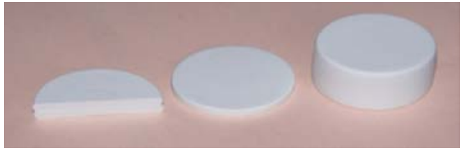
→ 3 compositions tested

| Supplier | Ref. | $SiO_2$ (%m) | $B_2O_3$ (%m) | $Al_2O_3$ (%m) | ZnO (%m) | CaO (%m) | Autres (%m) | CTE (25-300°C) (*$10^{-7}$ $K^{-1}$) | Melting point (°C) |
|---|---|---|---|---|---|---|---|---|---|
| Johnson Matthey | | 1* | 2* | 3* | | | BaO 4* | 62 | 920 |
| Emaux Soyer | REL063 | 1* | 2* | | | 3* | | 59 | 660 |
| Emaux Soyer | REL064 | | 3* | | 1* | | $Bi_2O_3$ 2* | 61 | 520 |

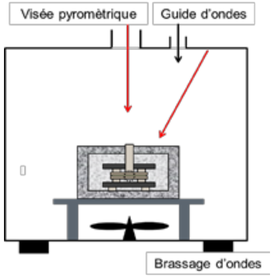
** 1 à 4 : sorting in descending order*

# Microwave heating for ceramic overpack sealing: Feasibility approach

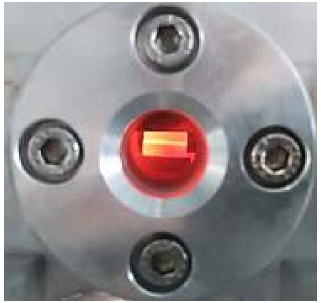
## First tests of microwave heating at laboratory scale

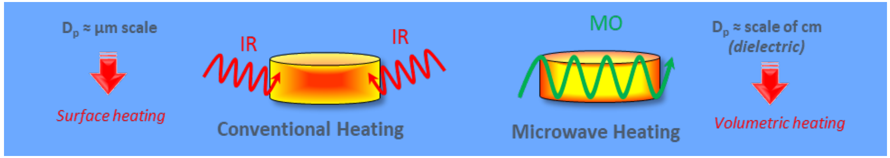



Aluminosilicate ceramic disk of 40 mm diameter and 5, 10 and 22 mm thickness



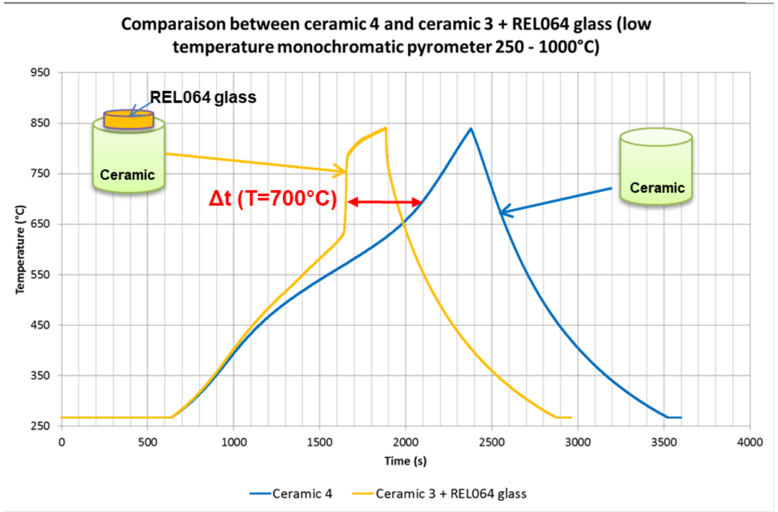

Microwave Furnace view

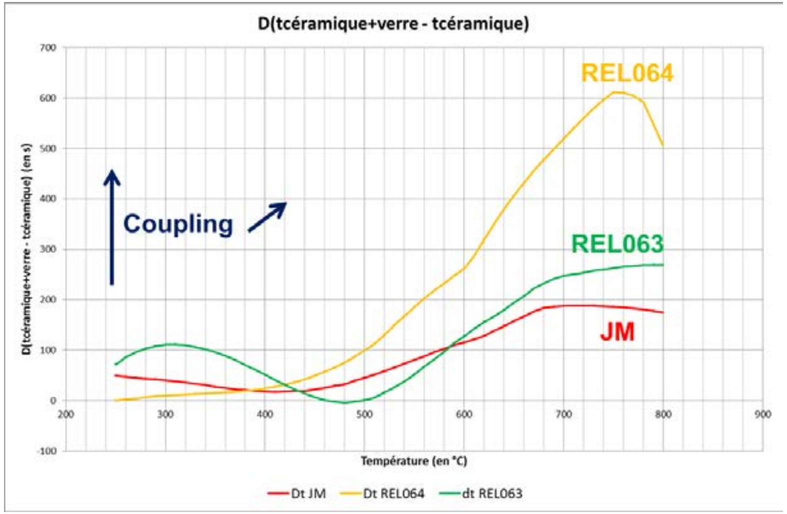





- Interaction between ceramic and microwave? Use of adding susceptor?
- Interaction between sealing glasses and microwave?

# Microwave heating for ceramic overpack sealing: Some results

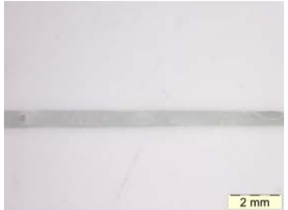


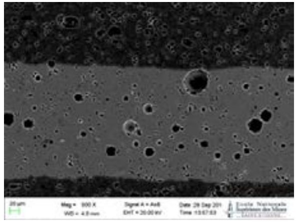




Direct microwave heating of ceramics

All glasses interact with microwave: REL63 < JM << REL64

# Microwave heating for ceramic overpack sealing: Some results



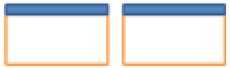
Feasibility of assembling by microwave heating achieved

Defaults (bubbles) observed in the glassy phases

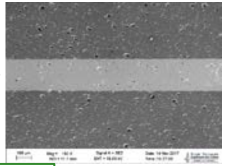
- o Curing of the enamel induce degassing
- o Bubbles are trapped in the assembly

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

Development of a two step heating process

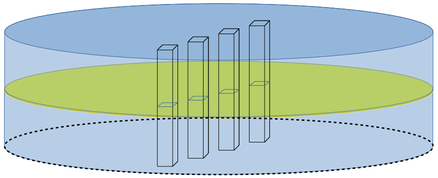


- o Enameling of the ceramic sealing surfaces (industrial process)
  - Spraying of the glass  slurry on ceramic
  - First heating of the enamel in a conventional furnace
- o Assembling of the enameled pieces by microwave heating



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 $mm^3$
4 point bending tests

## Breaking stress (MPa)

### Pencil brushed glassy slurry

| | REL 63 GLASS | REL 64 GLASS |
|---|---|---|
| Direct microwave joining | 15,7 ± 0,6 | 26,7 ± 5,2 |
| Two step process | 21,5 ± 2,0 | 76 ± 17 |

### Sprayed glassy slurry

| | REL 63 GLASS | REL 64 GLASS |
|---|---|---|
| Two step process | 147 ± 26 | 96 ± 34 |

*Ceramic : 135 ± 4MPa*

Improvement of the sealing quality with the new process





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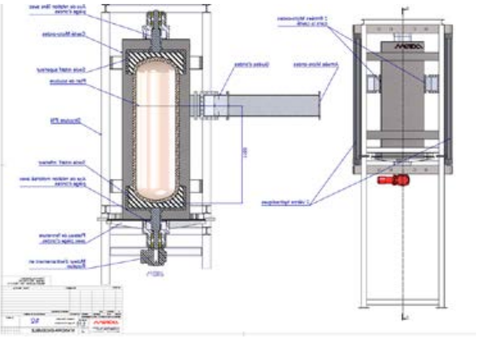
# Microwave heating for ceramic overpack sealing: **Next step…**



Is it possible to join ceramics pieces with an external microwave heating? (TRL 3 to TRL 4)

- Consulting of an industrial microwave furnace manufacturer (SAIREM)
- Development of a concept of microwave furnace for ceramic overpack sealing

Changing the scale and forms of the sample

- Ring forms
- ¼ and ½ scale

Development of a microwave furnace prototype to test ring assembling

Study of new sealing materials : ceramic-glass

- Better mechanical and lixiviation properties
- 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

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

## Sealing process

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





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# Thank you for your attention