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Influence of a thermal gradient and/or an electrical field on the crystallization of a piezoelectric phase in a glass-ceramic

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Piezoelectric ceramics

They are used in numerous components (sensors, actuators). These devices are polycrystalline and ferroelectric. They need to be poled under a high strength electric field before use. The principal drawback for these devices is the depolarization with time or increasing temperature. (Fig. 1)

Non-ferroelectric piezoelectric are not affected by this drawback but have lower electromechanical performances. Macroscopic polar properties can be conferred using glass-ceramic technology if a preferential orientation is promoting during crystallization step.

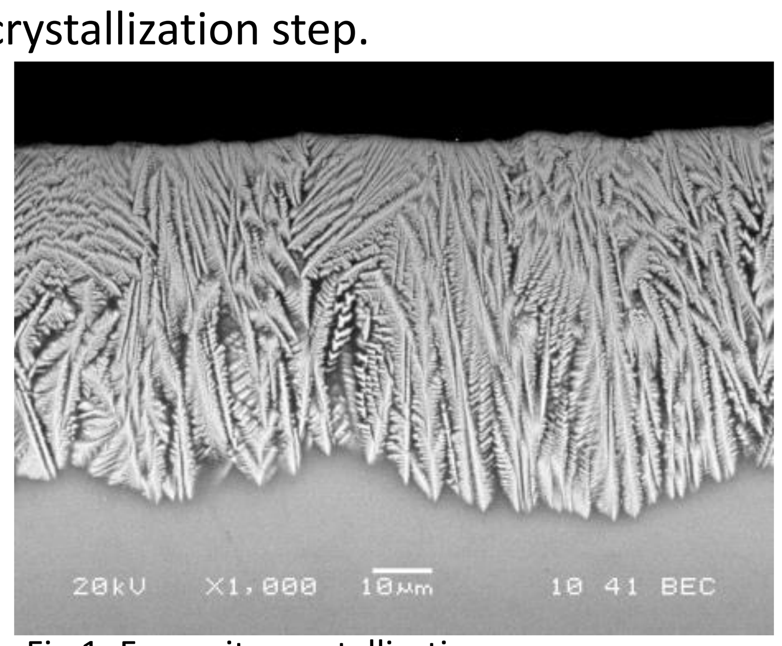


Fig.1: Fresnoite crystallization

N. Maury demonstrates in her PhD thesis[1], that a preferential orientation of piezoelectric fresnoite can be obtained by isothermal heat treatment on appropriated glass composition. The crystallization begins at the surface until the bulk. The crystallization mechanism promotes plans parallel to the surface: (001) lattice plans (Fig.1).

Glass-ceramic preparation

Glass-ceramic is prepared with strontium carbonate, potassium carbonate, silicium oxide, titanium oxide and alumina in stoichiometric proportion to eliminate phase separation: 3,3 SiO_2 1TiO_2 2 $SrCO_3$ 0,2 K_2CO_3 0,1 Al_2O_3. This composition was optimized during a previous PhD Thesis at UMONS[1].

Powders are stirred one night in isopropanol for homogenization (Fig. 2). The fusion of powders is made at 1550°C during 2 hours. The glass in fusion is poured onto a mold (Fig.3 and 4) and is placed 1 hour at 600°C. After, the sample is cooled slowly to room temperature to avoid cracks.


Fig.2: Powders Mix


Fig.3: Fusion

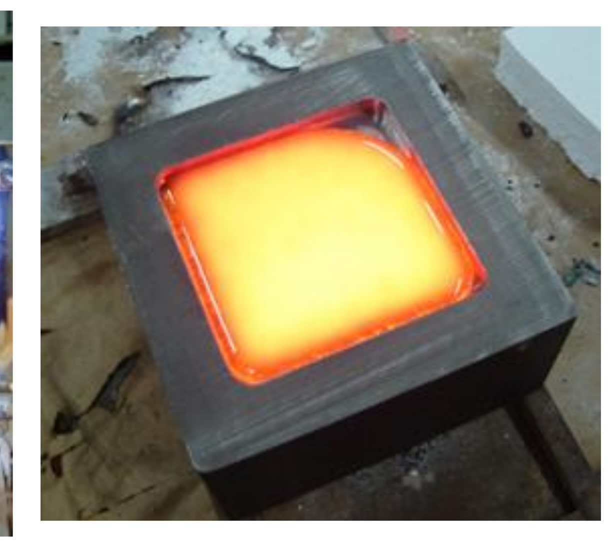


Fig.4: Glass tab

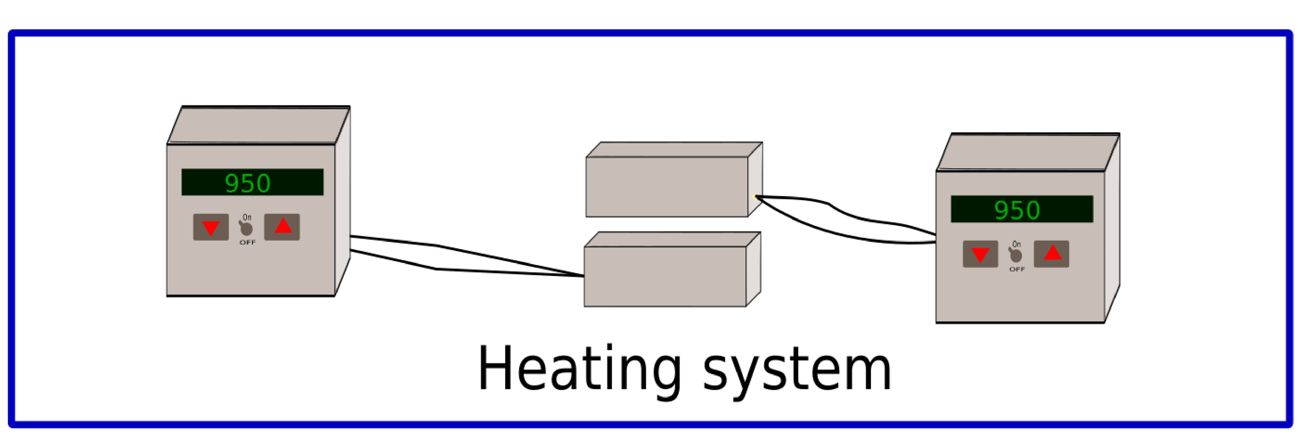
Glass-ceramics crystallization

The isothermal treatment gives modest piezoelectric performances because there is a loss of preferential orientation in the material bulk and a 180° dipolar moment inversions between crystals.

In this present work, to improve the glass-ceramic performances, a thermal gradient and a electric field [2,3] are applied during the crystallization.
In the first case, the aim is to favor crystals growth from the warmest side. In the second case, the aim is to force dipolar moments alignment. On that purpose a homemade furnace has been built.

The two independent heating sources are composed by:
- Refractory bricks
- Resistances, thermocouple and heat regulator
- Inconel plates
- Alumina tabs.

The Inconel plates are used for heat homogenization and as electrodes to apply the electrical field. The alumina tabs are used as electrical insulators (Fig.3).



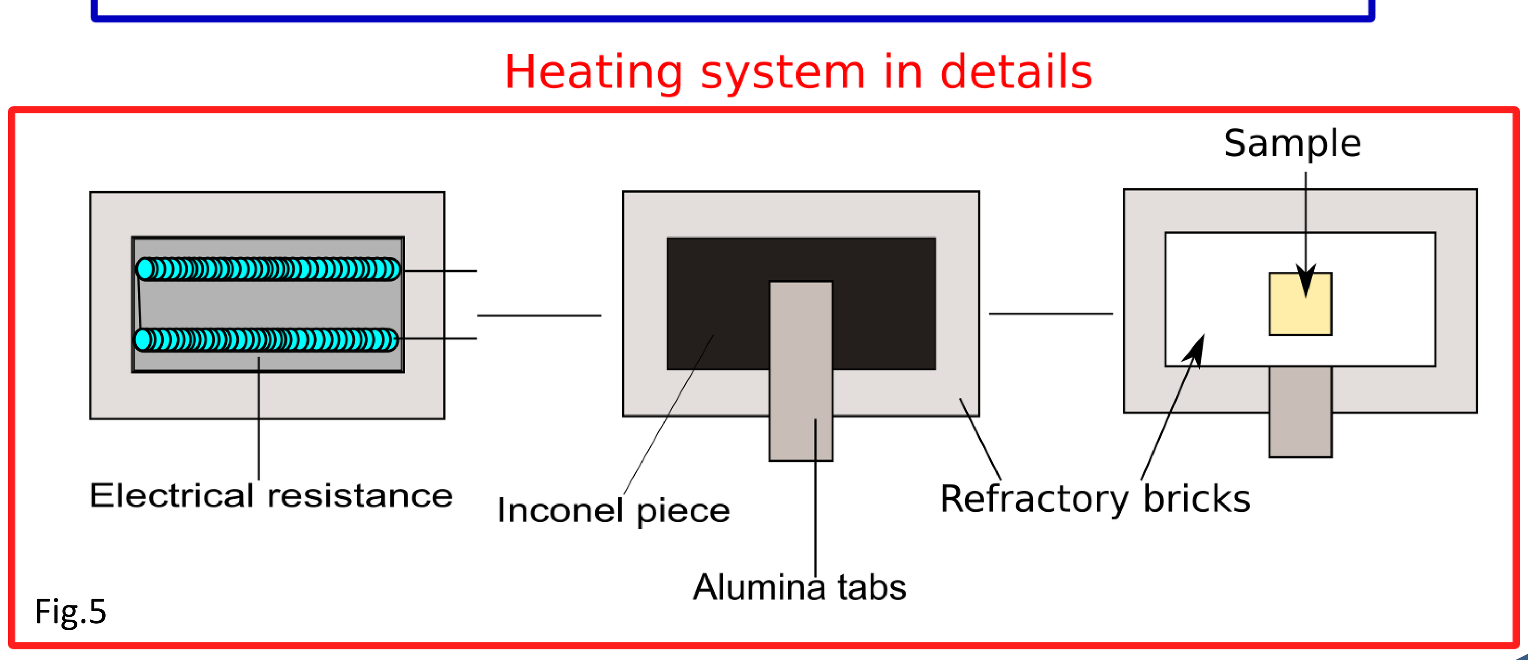


Fig.5

First results: thermal gradient

Crystallization observed during an isothermal treatment confirms N. Maury's results[1]: an orientated surface nucleation mechanism and a crystallization from the surface into the bulk (Fig.6 and 7).
In a convention furnace as with the homemade furnace in the absence of gradient, a lost of crystal orientation in the bulk is attested (Fig.6 and 7) and a misalignment of the crystals at the junction of the crystallization fronts is also observed (Fig. 8).

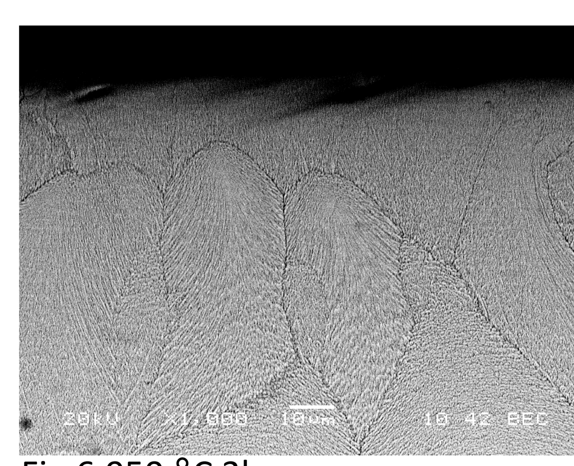


Fig.6:950 °C 3h

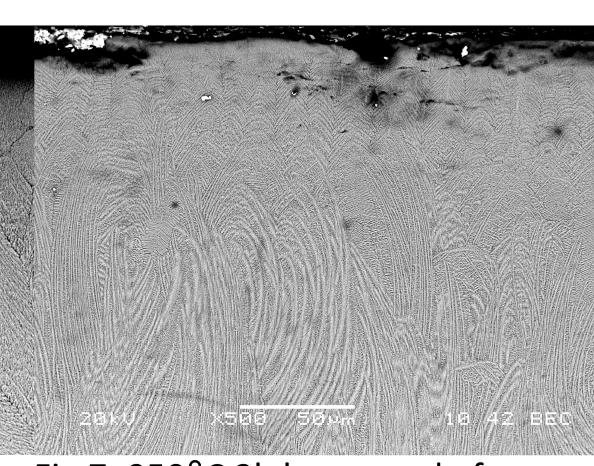


Fig.7: 950°C 3h homemade furnace

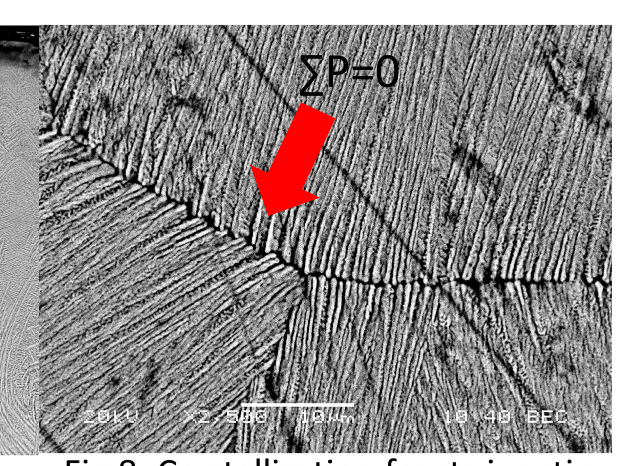


Fig.8: Crystallization fronts junction

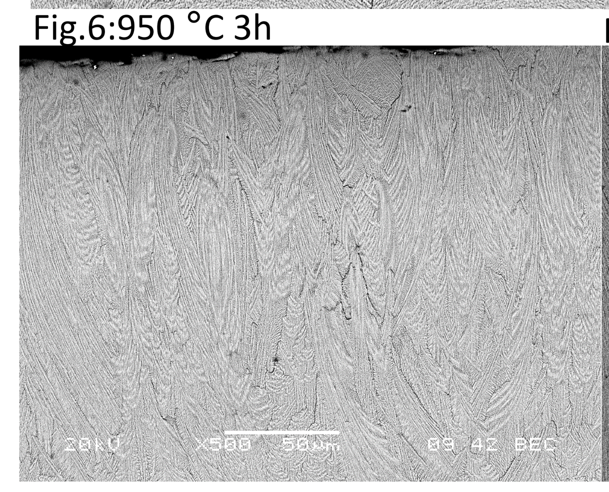


Fig.9:850°C/950°C- hot side

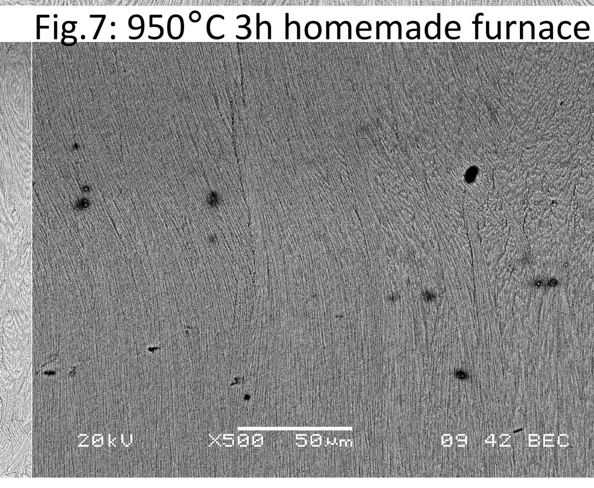


Fig.10: 850°C/950°C (-1 mm hot side)

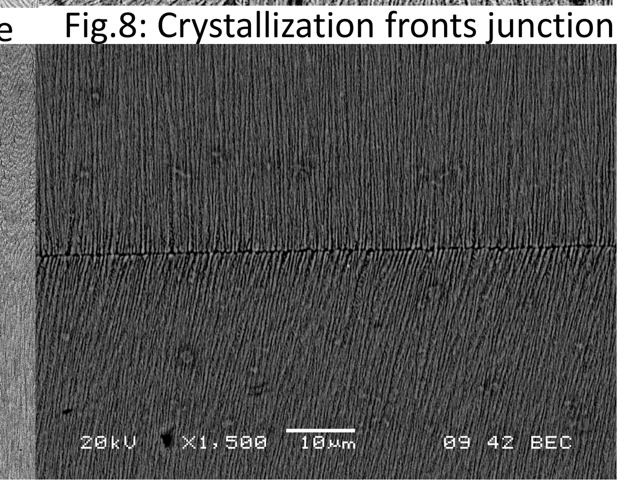


Fig.11: Junction point

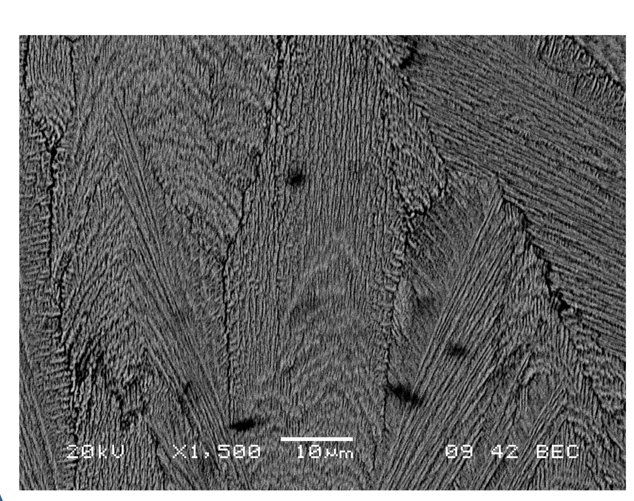
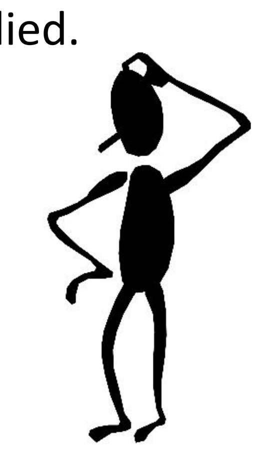


Fig.12: 850°C/950°C- cold side

On the opposite, when applying a thermal gradient of 100°C, crystal orientation seems to be better into the bulk (Fig 10 an 11) than near the hot (Fig. 9) or cold (Fig. 12) surface. As the crystallization rate is slower on the cold side than on the hot side, the junction of the crystallization fronts is not in the middle plane of the sample but is closer of the cold side (60 % / 40% of the sample thickness).

In comparison with an isothermal treatment, an higher preferential orientation of the crystal is obtained in the bulk when a thermal gradient is applied.
Following these first results, the upcoming tasks must be completed for all the samples :

- XRD analyses to characterize the crystals orientation and to follow its evolution from surface to the bulk
- Measurement of piezoelectric coefficient d_{33}
- Investigation of the effect of temperature setting (hot side, cold side, gradient)
- Investigation of the effect of an electrical field during crystallization treatment



[1] N.Maury, "Elaboration et caractérisation de vitrocéramiques piézoélectriques texturées à base de fresnoite", Thèse de doctorat UMONS, 2013.
[2] Y. Ochi, T. Meguro and K. Kakegawa ; « Orientated crystallization of fresnoite glass-ceramics by using a thermal gradient »; Journal of the European Ceramic Society 26 (2006)pp 627-630
[3] A. Halliyal, A.S bhalla, R.E. Newnham, L.E. Cross, T.R. Guruja; « Study of the piezoelectric properties of Ba2Ge2TiO8 glass-ceramic and single crystals »; journal of materials science 17 (1982), pp 295-300