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Densification and grain growth during sintering of porous $Ce_{0.9}Gd_{0.1}O_{1.95}$ tape cast layer

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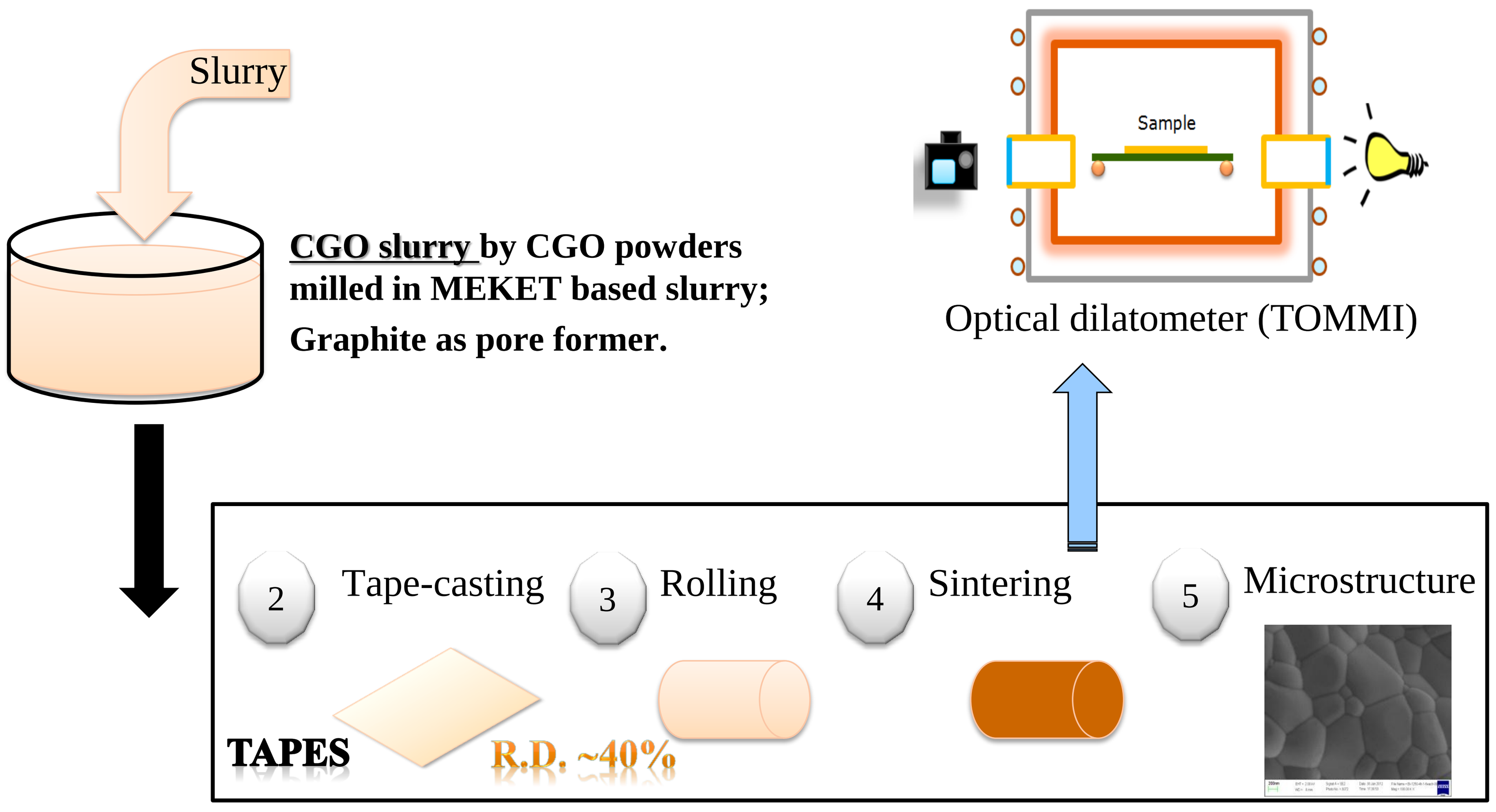
Motivation

The performance of ceramic devices is strongly dependent on its microstructure. In ceramic processing, sintering has a great impact on the final microstructures, since this step usually induces most of the morphological transformations at high temperature. Therefore, deep understanding of the sintering mechanisms and sintering kinetics is the basis for controlling the material microstructure and efficiently optimizing the ceramic properties. Moreover, the sintering mechanisms activation energy is one of the crucial input parameters for numerical sintering models. The sintering behaviour of dry pressed CGO pellet samples has been studied intensively. However, with respect to the CGO tape cast layer, a characterization is still missing. In this work, porous $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO10) tape with low-green density relevant for electrolytes in flue gas purification devices, was prepared by tape casting. Based on constitutive laws for densification, the sintering behaviour of CGO10 tape was systematically investigated to establish fundamental kinetic parameters associated to densification and grain growth.

Experimental procedure



Comparison of CGO10 pellet and porous tape

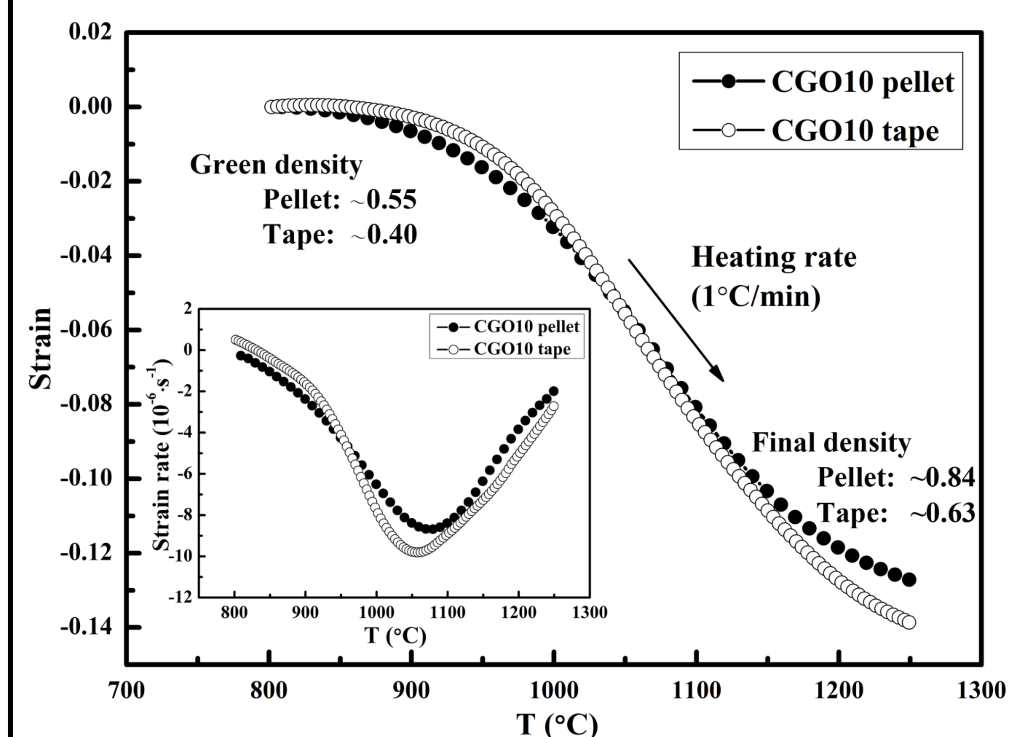


Fig. 1 Densification comparison of CGO10 pellet and porous tape as a function of sintering temperature.

Different densification behaviour between dry pressing pellet and porous tape

1. Effect of green density:
   - Pellets (0.55) and tapes (0.40)
2. Particle (pore) arrangement and anisotropy
   - Tape cast can obtain uniform microstructure with less agglomerates.

Accurate analysis of the sintering kinetics of tape cast layer cannot be carried out on pressed pellet samples and original shaped samples must be investigated.

Densification kinetics of porous CGO10 tape

Constitutive laws for densification:

$$-\frac{d\varepsilon}{dt}=\frac{1}{3\rho}\frac{d\rho}{dt}=\frac{C_1(\rho)\gamma D}{kTG^n}=\frac{C_1(\rho)\gamma D_0}{kTG^n}\exp(-\frac{Q}{RT})$$

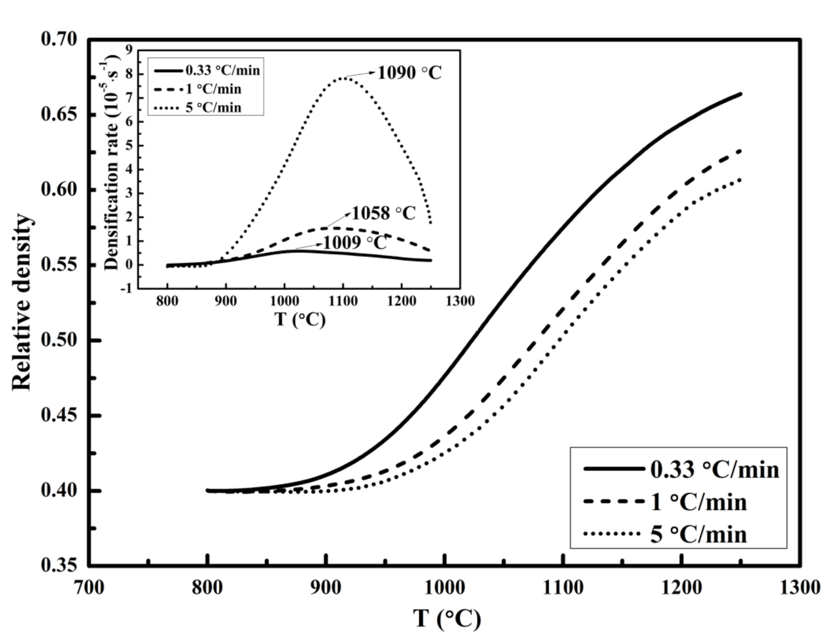


Fig. 2 Densification curves of porous CGO10 tape at different heating rates.

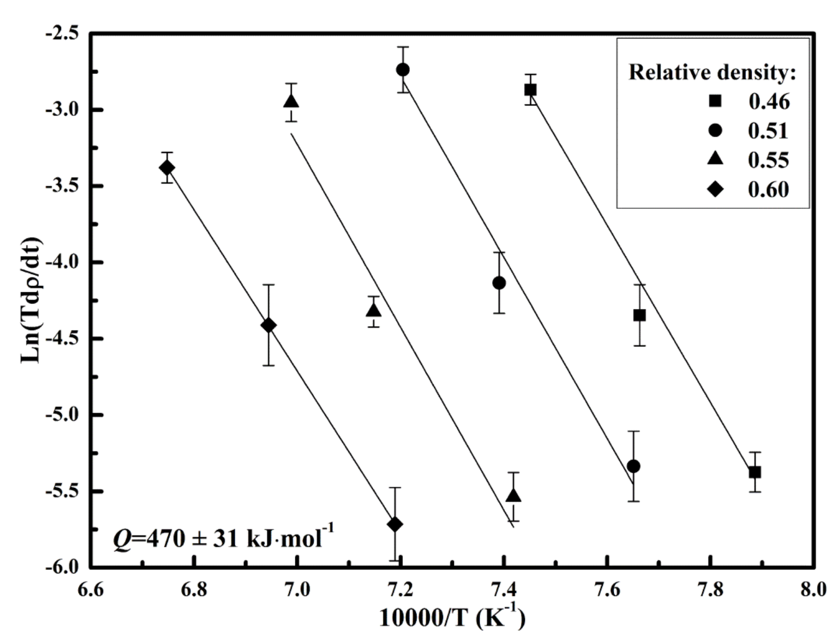


Fig. 3 Densification kinetics analyzed by using iso-density lines method.

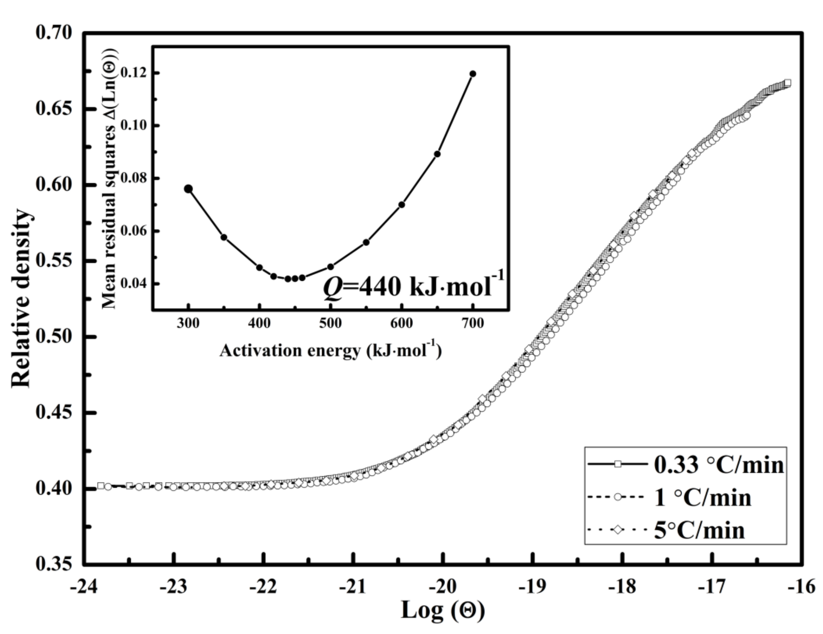


Fig. 4 Densification kinetics analyzed by using master sintering curve method.

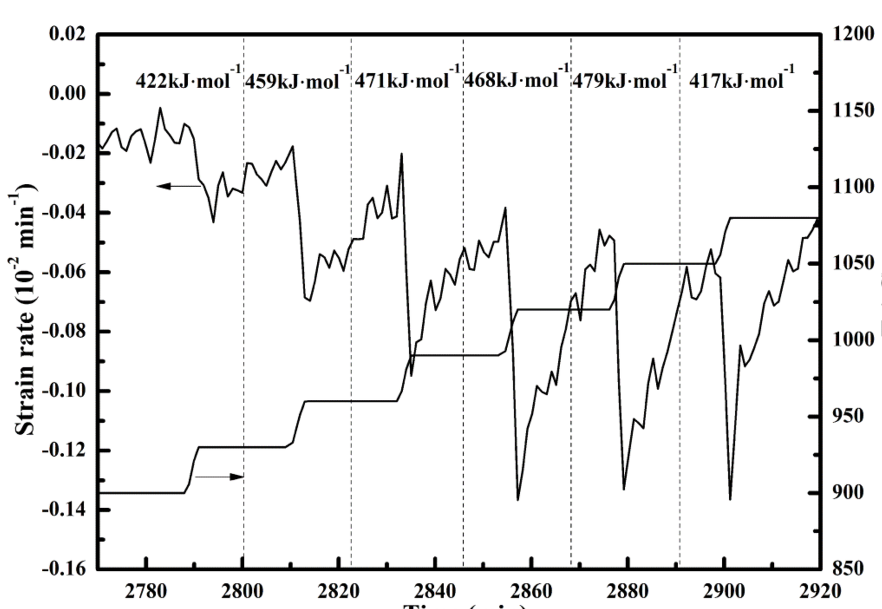


Fig. 5 Densification kinetics analyzed by using Dorn's method.

Grain growth kinetics of porous CGO10 tape

Kinetic laws for normal grain growth:

$$G_t^m - G_0^m = Kt = K_0 t\exp(-\frac{Q_g}{RT}) \qquad \Theta_g \equiv K_0 t\exp(-\frac{Q_g}{RT})$$

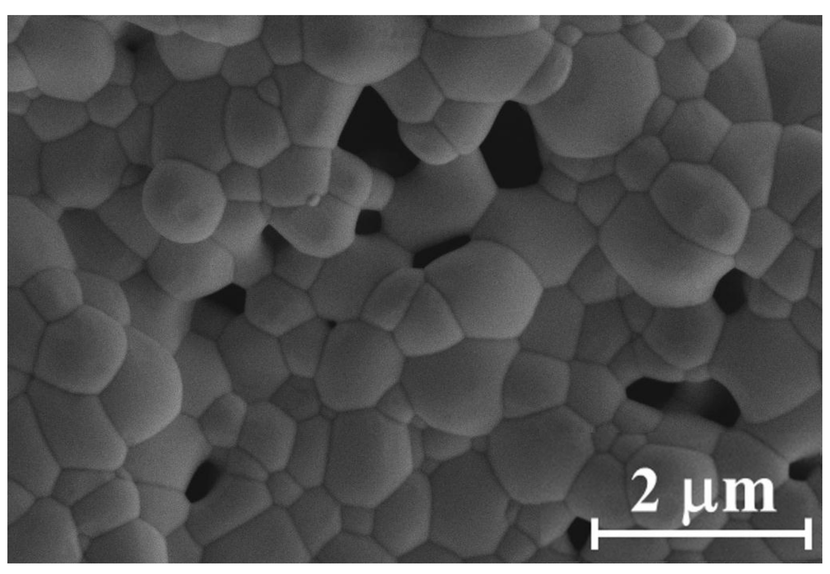


Fig. 6 SEM images of porous CGO10 tape sintered at 1250 °C for 4h.

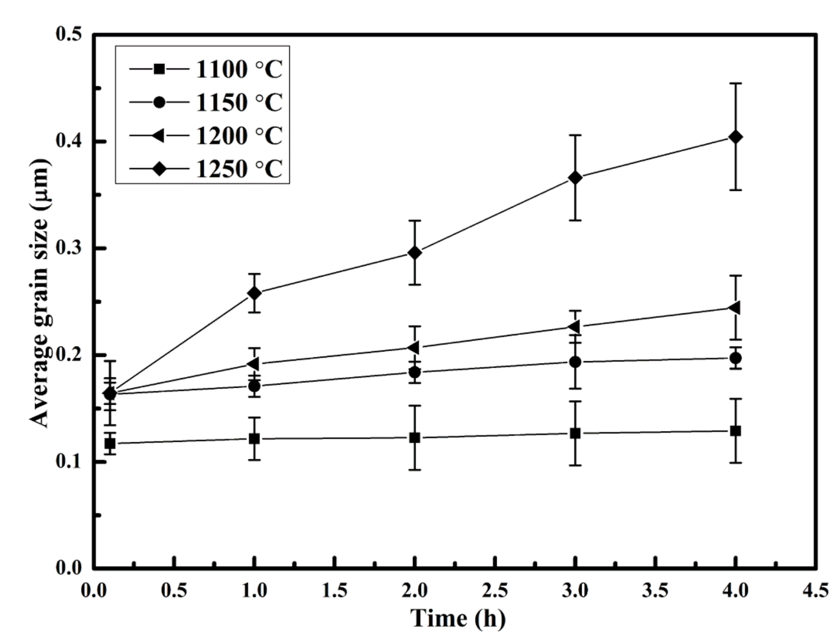


Fig. 7 Average grain size as a function of isothermal time at different temperatures.

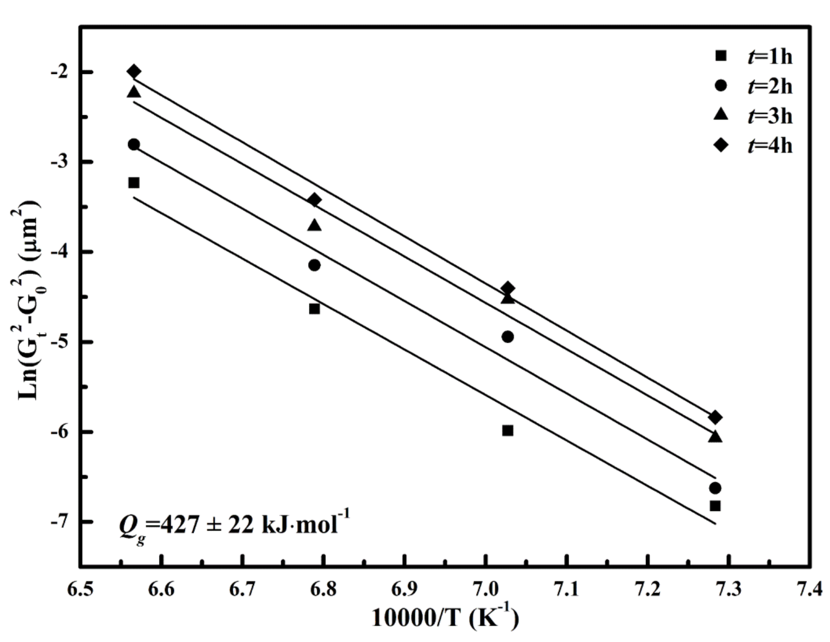


Fig. 8 Evaluation of grain growth kinetics for porous CGO10 tape.

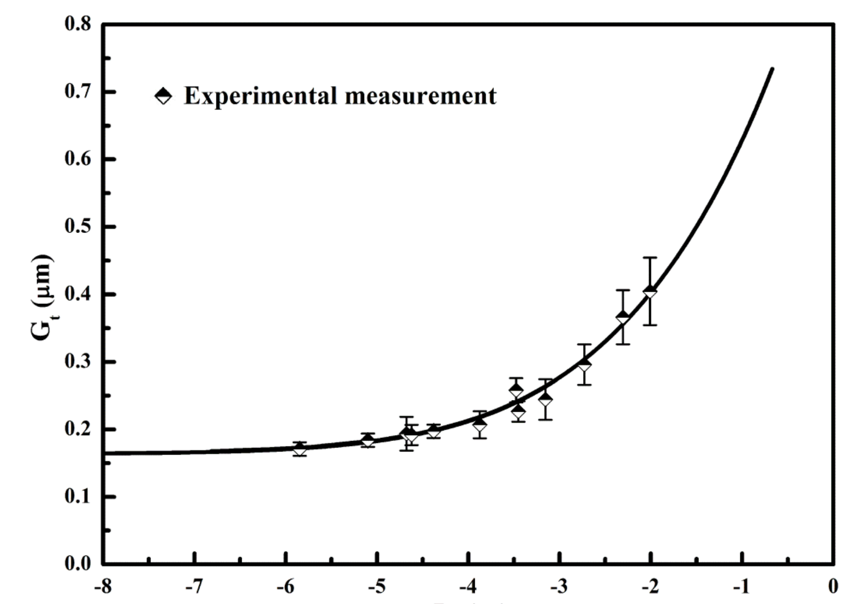


Fig. 9 Grain growth master curve for porous CGO10 tape.

Conclusions

- The densification behaviour of tape cast CGO10 layers is different from dry pressing pellet. Test a pressed pellet to characterize tapes can be misleading.
- The densification data were represented employing three different methods: iso-strain lines approach, master sintering curve (MSC) and Dorn's method, which produced similar kinetics results with densification activation energy of 440-470 $kJ\cdot mol^{-1}$.
- The grain growth activation energy of CGO10 tape was evaluated to be ~427±22 $kJ\cdot mol^{-1}$ and the grain growth master curve was constructed.
- It was indicated that the densification and grain growth processes for CGO10 tape were dominated by grain boundary diffusion mechanism and the grain boundary mobility was estimated around 10^{-18}-10^{-16} $m^3\cdot N^{-1}\cdot s^{-1}$ in the investigated temperature range.

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