Manufacture of cordierite parts by robocasting from recycled powders

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Abstract

This study deals with the formulation of a stable slurry of cordierite powder with a suitable rheological behavior for the robocasting process and with a sufficient solid content. The effects of 3D printing and sintering parameters on the printed parts regarding their final properties after firing such as density/porosity, expansion coefficient and Young's modulus are also investigated.

Keywords

Cordierite; Robocasting; Recycled powder; Rheology

Introduction

Advanced ceramics exhibit remarkable properties, such as high mechanical strength, thermal conductivity, or wear resistance. Therefore, they are used as critical components for specific applications in the fields of aerospace, automotive, energy production or cutting tools. Silicate ceramics usually present less efficient properties. However, they can be an economically advantageous alternative to technical ceramics in many applications operating at room or moderate temperature (< 1000°C). Moreover, regarding the environmental aspect, silicate ceramics are processed from natural abundant mineral resources, show a high recyclability, and require moderate sintering temperatures.

One potential technical use of silicate ceramics is the manufacture of gas cleaning devices, such as catalyst substrates or particulate filters, as it is the case for the cordierite in the automotive industry. The expansion of the use of these devices in other sectors goes through the development of low costs flexible manufacturing technologies and the improvement of the efficiency of the devices through the design components with complex architectures.

On that purpose, the aim of this study is to demonstrate the possibility to manufacture complex cordierite parts by robocasting. Moreover, this work focuses on the use of an already-formed cordierite powder as raw material, unlike the conventional route which starts from talc, kaolin, and alumina. The target is to pave the way to the use of recycled powders from industrial wastes.

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Results and discussion

Two cordierite powder were used. The first one (Powder I) is a recycled powder coming from a catalytic converter substrate (production waste without catalyzer). The powder undergoes milling in a steel ring mill. The second (Powder II) is a commercial powder (Tethon). This later was calcined at 600 °C for 1h to remove the possible organic additives. Both were characterized by X-ray fluorescence (XRF), X-ray diffraction (XRD), He-pycnometry and particle size analysis. The sinternability of these two powders was evaluated by means of pellets obtained by pressing under 50 MPa and fired at different temperatures and times. The robocasting pastes were prepared in deionized water with different solid loads. Different content of dispersing agent (Dolapic CE 64) and thickener (Rhodopol) were incorporated to control the rheological behavior. The rheology tests have been carried out with a Haake Mars III rheometer (Thermo Fisher Scientific) at temperature room using a parallel-plate geometry (PP20Ti, gap of 1 mm). The suitable pastes were used to print dense parts using a LYNXTER S600D. The printed parts were dried in a climatic chamber and then sintered. The Young's modulus of the sintered parts was measured by Impulse Excitation Technique (Grindosonic) and the thermal expansion coefficient by means of a dilatometer.
The XRF confirms the composition of cordierite (Table I) but XRD highlights some intermediate compounds in the formation of cordierite, such as mullite, in the case of the commercial powder (Fig. 1). This can explain the difference in density (Table I). The particle size distribution of the two powders is similar (Fig. 2)

Table I: Equivalent-oxide composition (mol%) of the powders from the XRF analysis and absolute density (g/cm^3) from the He-pycnometry

	Al_2O_3	SiO_2	MgO	Density
Powder I	21	56	22	2,60
Powder II	24	54	21	2,70

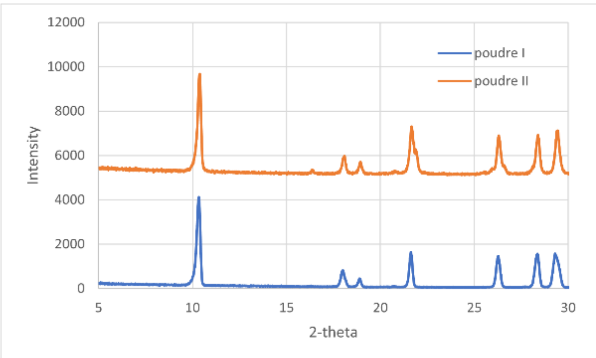

Fig. 1.: XRD patterns of the powders

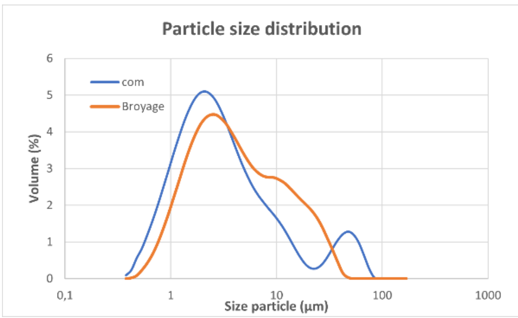

Fig. 2.: Particle size distribution

The study of the sinterability highlights that densification begins at 1200 °C (Fig. 3). The commercial powder shows a higher porosity due to its lower green density. During an isothermal sintering at 1300°C a decrease in porosity is observed with the time for the commercial cordierite (Fig. 4).

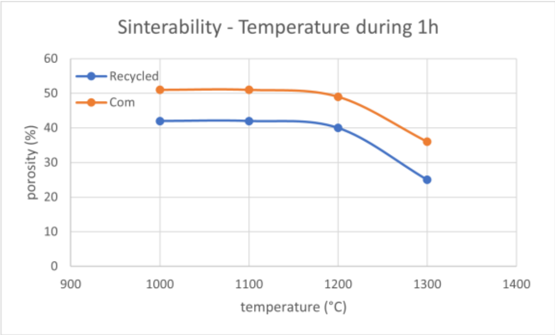

Fig. 3.: Effect of the temperature

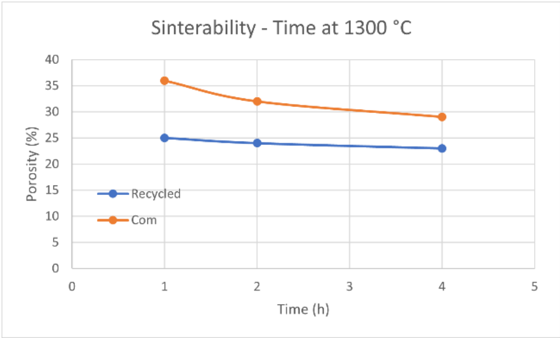

Fig. 4.: Effect of the time

With the objective to increase the solid load, the optimum amount of dispersing agent has been found from the lowest viscosity at the shear rate obtained during the printing; 1,25 wt% for both powders (Fig.5). With this amount, it was not possible to load the paste over 45vol% for the commercial powder, whereas 57.5vol% was reached with the recycled powder. However, the yield stress was null, and then the printed paste crushed down during the printing.

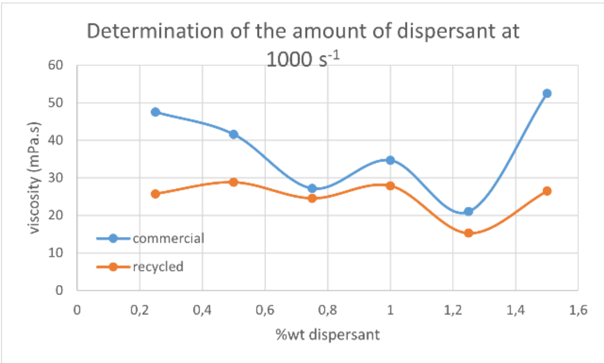

Fig. 5. Influence of the amount of dispersing agent on the viscosity of the paste

To avoid this problem, a thickening agent (Rhodopol) has been added to the paste in different concentrations (/wt of water). The optimum has been found by measuring the yield stress and yield flow by an amplitude sweep test (Table II) and the difference between the STL file and the printed part (Fig 6). The optimal concentration was 4,76%wt of Rhodopol for both powders.

Table II: Influence of concentration of thickening agent (Rhodopol) on the yield stress (Pa) and the yield flow (Pa) measured by amplitude sweep test.

Sample	Powder I			Powder II	
	Yield stress	Yield flow		Yield stress	Yield flow
0,59R	4	6		141	433
1,19R	10	35		75	166
2,38R	24	143		63	378
4,76R	87	825		39	612

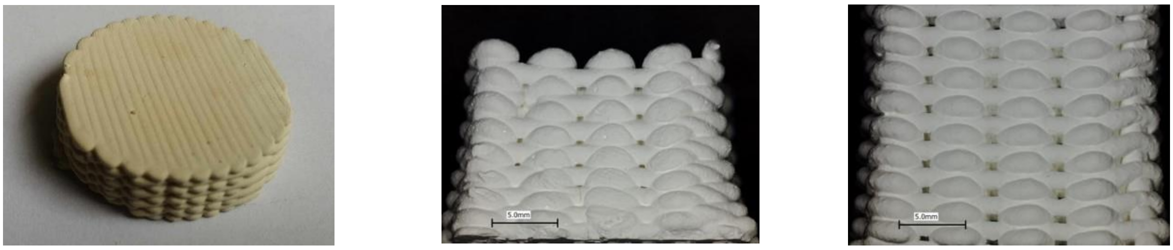

Figure 6: Images of the printed parts. From left to right: Powder I 4,76R; Powder II 2,38R and 4,76R.

The printed parts have been dried and sintered at 1300°C for 4h. The porosity of the samples has been calculated by Archimedes principle. For the recycled powder, the total porosity was 23% with 21% of open porosity. For the commercial one, the total porosity was 28% with 22% of open porosity. These results are close to those obtained with the pressed powders (Fig. 4).
Mechanical characterization has been achieved by measuring the Young Modulus by Impulse Excitation Technique. The Hashin-Shtrikman model has been applied to the results to avoid the influence of the porosity. For the pressed samples the Young's moduli are 117 GPa for powder I and, 147 GPa for powder II. For the printed samples, the Young's moduli are respectively 76 GPa and 58 GPa.
Finally, the thermal expansion coefficients (CTE) have been measured by dilatometry and compared to those of an extruded honeycomb substrate following the longitudinal and transversal orientation of the extrusion (Table III). For the commercial cordierite, a pressed and sintered sample was also characterized as a reference. For the printed parts, CTE has been measured following the axe Z. The results are reported in the table below.

Table III.: CTE of the samples

Sample	Honeycomb (Powder I)	Powder I printed	Powder II pressed	Powder II printed
CTE (10^{-6} K^{-1})	0,8 – 1,7	1,8	2,5	2,7

Conclusions

Cordierite parts have been successfully printed from a recycled powder and a commercial powder. The rheology behavior of the pastes has been optimized by the amount of dispersing agent and the use of a thickener. After sintering, the Young's modulus of the printed parts is lower than that the pressed parts, what can be due to the presence of defects. The thermal expansion coefficient of the parts printed from the recycled powder is very close to that of the initial honeycomb substrate. Future studies will focus on the enhancement of the mechanical properties.