Continuous and Contactless Method of Measuring Shrinkage and Temperature Distribution during Microwave Sintering

Daniel Zymelka, Sébastien Saunier, Jerome Molimard, and Dominique Goeuriot

Département Mécanique et Procédés d'Elaboration, Centre Sciences des Matériaux et des Structures, Ecole Nationale Supérieure des Mines, Saint-Etienne, France

A contactless technique for measuring the evolution of physical and microstructural parameters during microwave sintering using a charge-coupled device (CCD) camera and infrared (IR) pyrometers will be shown. The purpose of this study is to follow the temperature field and the shrinkage of typical ceramic parts during microwave sintering. Temperature measurement using a CCD camera is based on Stephan-Boltzmann's law. The mean temperature and pixel intensity values were registered at the same time and on the same area by infrared pyrometers and a 12-bit CCD camera respectively. Based on these two values, the relationship between intensity and temperature was determined. Simultaneously the distribution of pixel intensity over the whole surface of the sample was registered and converted to temperature. The shrinkage measurement was done using a boundary detection algorithm. The experiments were executed in real time. This technique will be applied to determine sintering activation energy of different ceramic materials during microwave heating.

Introduction

Dimensional variation measurements and the kinetics of this process with simultaneous measurement of temperature are an essential part of the investigation of sintering mechanisms.

To measure temperature of materials undergoing conventional heating, thermocouples are commonly used. However, during microwave heating electromagnetic interference problems may occur, due to the necessity of inserting a thermocouple into the cavity. Applying IR pyrometers, in spite of problems with calibration because of different values of emissivity for different materials, allows contactless measurement, which should make it more effective for use in microwave processing.

During microwave sintering, non-homogeneous microstructure of materials can occur due to the development of heat distribution. Monitoring the temperature field can give us important information which may be useful for observing the evolution of the thermal gradient and defining its scale. This can be important for investigating the densification process which can be blocked by the thermal gradient [1].

In conventional sintering, the shrinkage measurement is usually conducted using a dilatometer to measure the position variation of a mobile aluminum piece which has to be in contact with the sample. Because of technical difficulties, shrinkage measurements are not commonly used during microwave sintering. However, Marinel *et al*. [2] have presented interesting results of shrinkage measurements versus time in single mode microwave heating. But we should pay attention to the fact that owing to the small dimensions of single mode cavities, in these systems it is possible to use a light element to measure dimensional variation. The use of the same method in multimode cavities requires the use of a longer, and thus heavier, element. This in turn may affect the densification process. To avoid this situation, we would have

to use a standard pushrod dilatometer with controlled force of pressure, which is very difficult. Furthermore an electromagnetic interference problem may occur.

For multimode microwave cavity we propose a contactless and continuous method using a CCD camera. Thanks to this method the pellets' dimensional variation versus time and temperature distribution on the sample's surface can be monitored.

Technique

To measure the temperature an IR pyrometer is used. A 12-bit CCD Camera with 2k x 2k matrix registers 2 images per second. The frequency of data recording can be changed as needed. The lens of the camera has been chosen for samples that are 20 mm diameter. The process is controlled by a dedicated program, and is repeated with constant frequency. A 430 x 430 x 490 mm^3 multimode cavity and 2.45 GHz generator have been used. Green samples of ZnO were prepared by uniaxial pre-compaction at 80 MPa into pellets followed by isostatic press at 400 MPa. The green densities were 67% TD.

Temperature measurement

The aim here is to detect in real time, during the experiment, a complete temperature field. It is expected that during the heating process, some temperature variations (hot spots) will occur. The temperature measurement is based on data obtained using the CCD camera and real-time calibration with a punctual pyrometer. The measurement area of the pyrometer depends on the distance between the pyrometer and the object. The mean value of intensity and temperature at the same time and over the same area are measured (Fig. 1A). Based on this data, the relationship between temperature and intensity is plotted in real time (Fig. 1B), and a simplified exponential law is fitted. Then, each of the CCD matrix pixels can be converted to temperature values according to equation 1:

$$Temperature = \ln\frac{(Intensity\ )}{(Amplitude\ )} \cdot (Damping)^{-1}$$

Note that during the experiment, the settings of the camera and lens were not changed. Moreover, all automatic settings of the camera (white balance, gain, exposure, etc.) should be switched off.

Using our CCD camera we can register thermal emissions only in the visible and near-infrared ranges. The intensity reaches a sufficient level at approximately 600°C. The pyrometer using infrared emission is sensitive to lower temperatures, from 250°C. Note that the camera acts as an integrator over a certain wavelength range of Stephan-Boltzmann's law, with each wavelength being weighted by the CCD response [3].

A complete uncertainty analysis is still impractical due to our lack of experimental background on the device, but some key points can be given anyway. Fig. 2A shows uncertainty of the temperature field when using our 12-bit CCD camera. For comparison, the results of temperature field uncertainty when using an 8-bit CCD camera are presented in Fig. 2B. It is shown that using a 12-bit camera can yield results with a lower error than using an 8-bit camera. We can also see that the error decreases with the temperature.

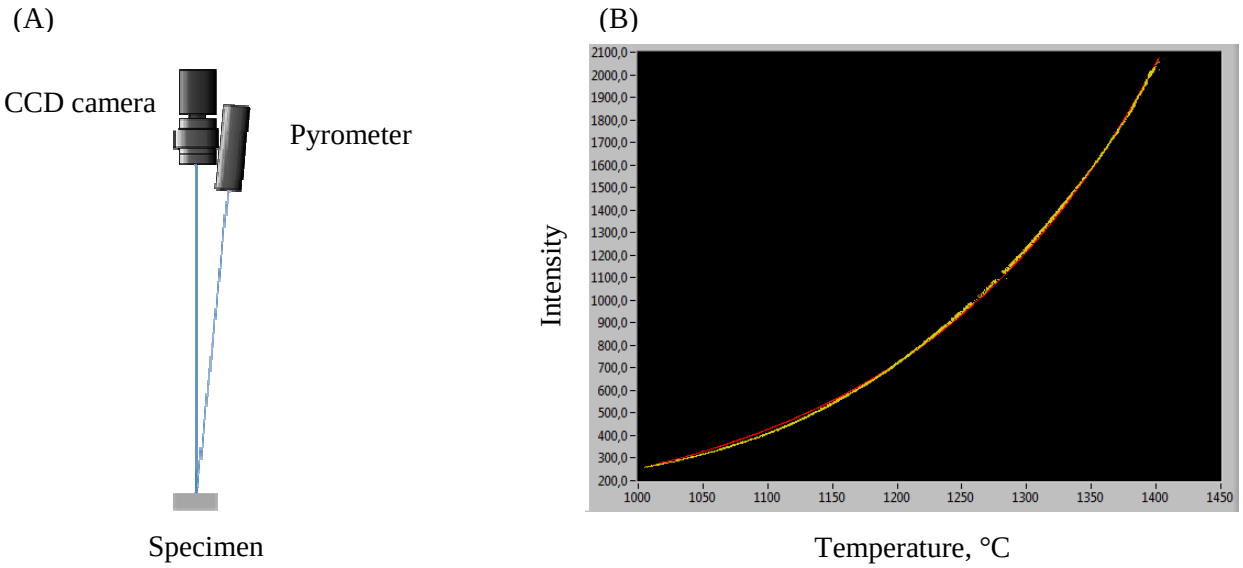

Fig. 1. (A) Scheme of the optical system used during the experiment. (B) The yellow line represents the experimental data; the red curve was calculated in real time based on Equation 1.

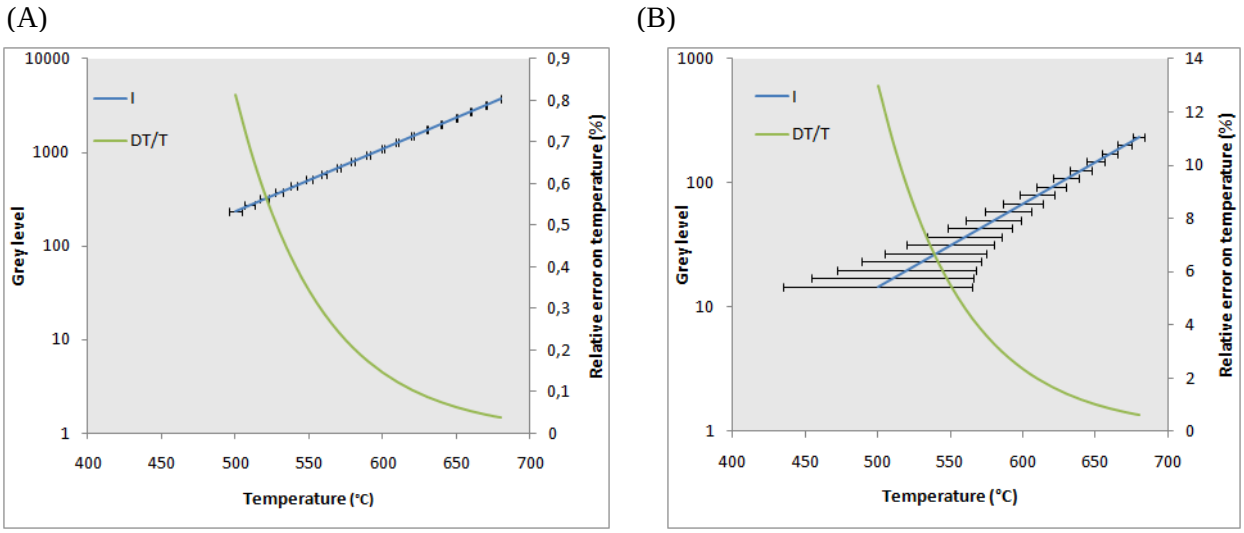

Fig. 2. (A) Uncertainty of temperature field when using our 12-bit CCD camera. (B) Uncertainty of temperature field when using an 8-bit CCD camera.

Shrinkage measurement

Realization of this process is based on the contrast between the specimen and the environment. Again, shrinkage is estimated in real time at every image acquisition step.

At the beginning the 12-bit image was recorded; however, between registering a basic image and the last step, which is the diameter measurement, the image has to be prepared for calculations. This is the most difficult step because we cannot use any external light or camera automatic exposure which can improve the contrast, due to measurements of temperature field. Basic operations are threshold and a non linear low-pass filter to remove erroneous points. Next the boundary detection algorithm is applied. In this work we have used an algorithm constructed

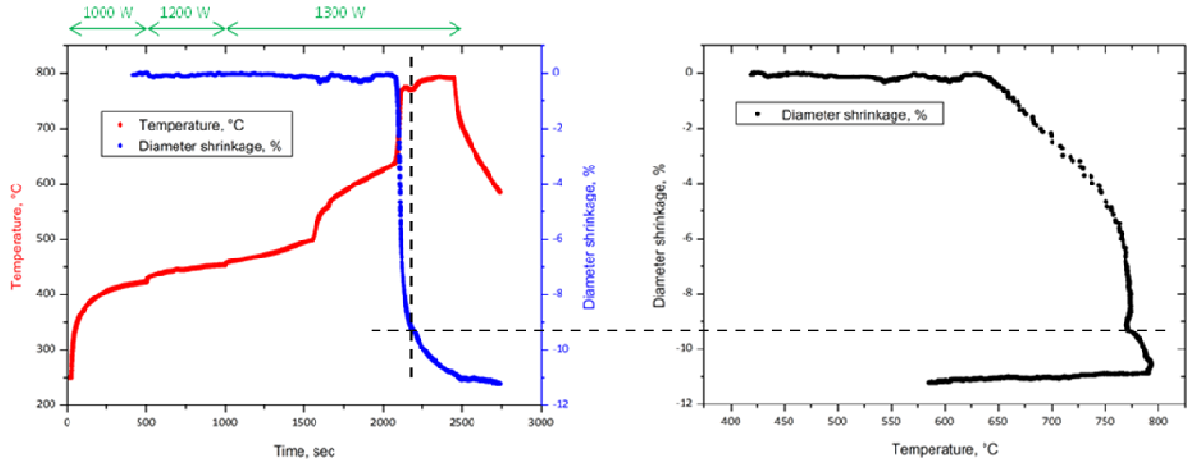

Fig. 3. Shrinkage curves versus time and temperature of ZnO sample heated by microwaves.

by National Instruments. The algorithm searches a circular edge in an area of exploration, and locates the intersection points between defined lines and edges of the object based on the contrast.

At the end, the diameter of the sample is calculated based on the founding edges, and is presented in the graph as a function of time and temperature in real time.

The precision of dimensional variation measurements strongly depends on the image sharpness. To increase the precision of measurements, the maximum surface of the CCD matrix should be used.

Results

In Fig. 3 the dimensional variation of ZnO is shown. We observe that densification occurred very quickly, but this is likely due to thermal runaway at approximately 700°C. In the initial part of the curve presented in Fig. 3, we observe an abnormal trajectory of the dilatometric curve. This issue is likely caused by the algorithm's having detected the wrong edges of the sample. Note that at the beginning of heating, the algorithm operation was carried out with poor light intensity. This problem will be considered in the coming work.

Before and after sintering, the diameter of the sample was measured manually using a caliper. After that, the results were compared with those obtained using the optical method (Fig. 3). The diameter variation measured manually is 10.92%, whereas the diameter measured by the CCD camera is 11.08%. The final density of ZnO pellets is 97.7% TD.

Fig. 4 shows the evolution of the temperature field on the surface of the ZnO sample. We can see clearly that during direct microwave heating the temperature distribution is increasingly heterogeneous. The value of the gradient developed can reach 100°C/cm, which in turn can affect the densification process.

Conclusion

We have presented an innovative method of exploring the microwave sintering process. This method is still under development. In future work we aim to use it to investigate sintering

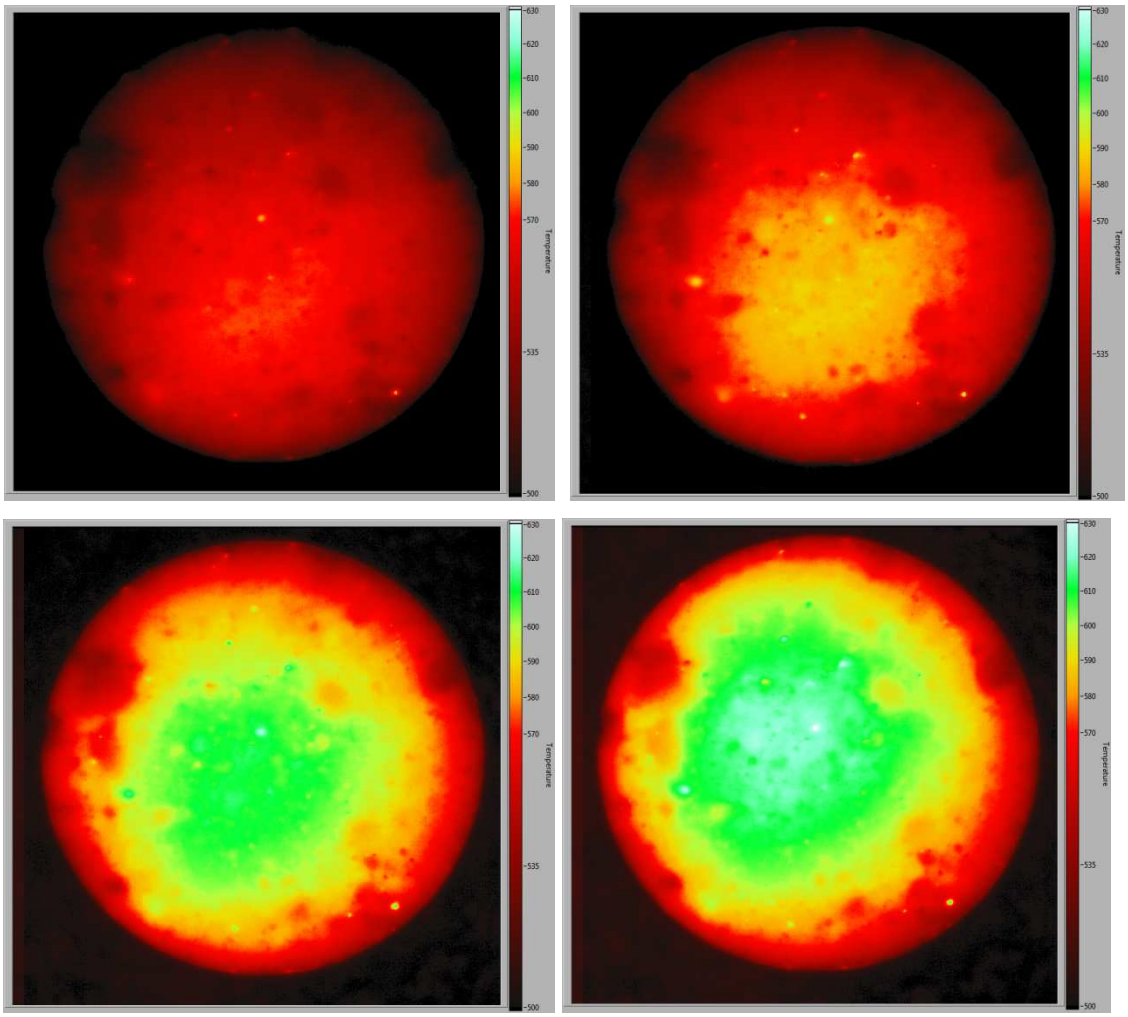

Fig. 4. Evolution of the temperature field on the surface of the ZnO sample.

mechanisms during microwave heating. We also believe that this method may give important information which might be useful to quantify the effect of hot spots and thermal gradient developed during microwave sintering.

References

[1] S. Saunier, F. Valdivieso, Modification of mass transport during sintering induced by thermal gradient, *Advanced in Sintering Science and Technology*, Ceramic Transactions, vol. 209
[2] S. Marinel, E. Savary, In situ measurement of the shrinkage during microwave sintering, *Journal of Materials Processing Technology*, Issue 10, pp. 4784-4788, 2009.
[3] T. Sentenac, Y. Le Maoult, G. Nsengumuremyi, Y. Rotrou, G. Bernhart, Mesure de champs de température par caméra CCD : application au suivi thermique de moules, *École des Mines d'Albi.*