Influence of technological parameters on the hot-pressed process of CaF_2 optical ceramics

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Received 28 Mar. 2025; Revised 19 May 2025; Accepted 10 Aug. 2025; Published 25 Aug. 2025.
DOI: https://doi.org/10.54939/1859-1043.j.mst.105.2025.113-120

ABSTRACT

The paper uses the experimental planning method to study the influence of technological parameters on the hot pressing process of CaF_2 optical ceramics. The experimental planning model is applied to build the correlation equations between technological parameters, such as temperature, time, and pressure, on the quality index of the transmittance of optical ceramics. From there, the authors determine the optimal set of hot pressing technological parameters for the hot pressing process of CaF_2 optical ceramics. The research findings serve as an important basis for further studies on the hot-pressing process of various optical ceramics used in military optical devices.

Keywords: Experimental planning; CaF_2 optical ceramics; Hot-pressed process; Transmittance; Optical equipment.

1. INTRODUCTION

Calcium fluoride (CaF_2) optical ceramics are promising materials for high-performance infrared (IR) systems due to their excellent IR transmittance, corrosion resistance, thermal shock tolerance, and mechanical strength. These properties make CaF_2 a strong candidate for components such as lenses and windows in IR tracking and guidance applications, particularly within mid-wave infrared (MWIR) systems [1–3]. In recent studies, CaF_2 optical ceramics have demonstrated promising performance in infrared applications, particularly in the MWIR range. For instance, sintered CaF_2 ceramics have achieved transmittance levels exceeding 80% at 4.5 µm wavelength, which meets the optical quality requirements for advanced tracking and imaging systems. These properties not only validate the theoretical advantages of CaF_2 but also confirm its practical potential in real-world defense optics. Furthermore, its stable performance under high thermal and mechanical loads has positioned CaF_2 as a viable alternative to single crystals in many infrared systems. These encouraging results underscore the importance of continued research on optimizing the processing parameters to maximize the material's optical quality and mechanical durability.

Currently, hot-pressing is one of the most common and widely researched technologies for producing optical ceramics, CaF_2, around the world [4-6]. Using hot-pressing techniques, various infrared optical ceramics, such as Irtran-1, Irtran-2, or KO-1, KO-2, KO-3, KO-1, have been successfully fabricated. Currently, in our country, CaF_2 optical ceramics and the hot-pressing technology for manufacturing optical ceramics remain relatively new and have not been thoroughly researched [7, 8]. Therefore, this paper investigates the influence of technological parameters on the hot-pressing process of CaF_2 optical ceramics, which is essential for advancing the field.

To address the above challenges, this study is structured as follows: section 2 introduces the materials, equipment, and experimental procedures used in the hot-pressing process of CaF_2 optical ceramics. Section 3 presents the experimental results, regression modeling, and statistical analysis to determine the relationship between the technological parameters and the optical transmittance. The discussion also includes model validation and optimization of hot-pressing conditions. Finally, section 4 concludes the study by summarizing key findings and outlining directions for future research.

2. EXPERIMENTAL

2.1. Research object

The material used in this study is CaF_2 nanopowder with specifications given in table 1.

Table 1. *Specifications of CaF_2 nanopowder material.*

Purity (%)	> 99
Particle size (nm)	80 ÷ 100
Melting point (°C)	1420

To confirm the chemical composition of the CaF_2 nanopowder used in this study, Energy Dispersive X-ray Spectroscopy (EDS/EDX) analysis was conducted. The EDX results, shown in figure 1 and table 2, indicate the presence of calcium (Ca) and fluorine (F) elements with approximate atomic percentages of 33.90% and 66.11%, respectively. These values are consistent with the theoretical stoichiometry of CaF_2 (1:2 ratio of Ca to F), thereby confirming the chemical purity and suitability of the material for optical ceramic applications.

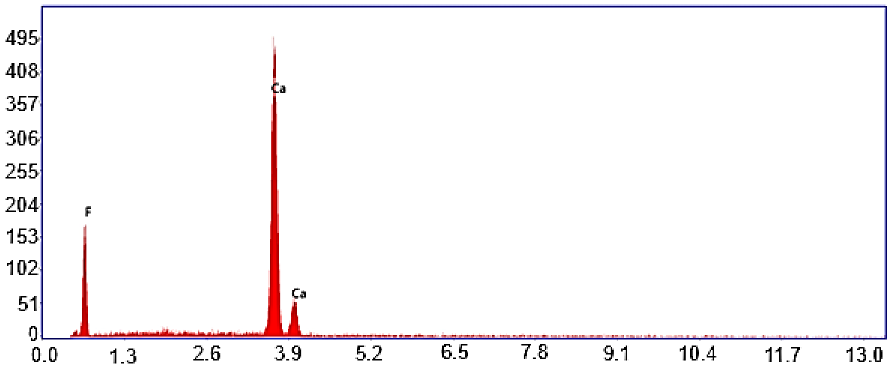

Figure 1. *EDX spectrum of CaF_2 nanopowder.*

Table 2. *Elemental composition of CaF_2 nanopowder.*

Element	Weight %	Atomic %	Net Int.	Error %	Kratio	Z	R	A	F
F K	48.04	66.11	240.12	11.62	0.09	1.04	0.96	0.19	1
CaK	51.96	33.90	841.45	4.12	0.50	0.96	1.03	0.99	1.01

2.2. Methodology

The experimental design focuses on evaluating the influence of three key processing parameters: temperature (T), holding time (t), and applied pressure (P). These factors were varied systematically based on a central composite design to explore their individual and interactive effects on the optical transmittance of CaF_2 ceramics. Other processing parameters, including heating/pressing rate and atmosphere, were fixed to eliminate confounding effects.

To reduce the number of experiments while still capturing the effects of each factor, we employed a statistical experimental design approach. This method saves time and resources by constructing a mathematical model (regression equation) to describe the experimental behavior instead of performing all possible experiments. The model is chosen as a polynomial that can approximate any arbitrary function, with the degree of the polynomial determining the accuracy of the model. Higher-degree polynomials can improve the model's accuracy, but one must balance accuracy against increased complexity. A general second-degree (quadratic) regression model [9, 10] for three factors can be written as:

$$f = b_o x_0 + b_1 x_1 + b_2 x_2 + b_{(k-1)k} x_{(k-1)} x_k + b_{11} x_1^2 + b_{22} x_2^2 + \ldots + b_{kk} x_k^2 \qquad (1)$$

Where: f – Objective function, representing the modeled relationship being studied; x_i – Factor affecting the objective function; b – Regression coefficient;

The absolute value of each regression coefficient b reflects the degree of its influence on the objective function. A smaller absolute value indicates a weaker effect, whereas a larger value indicates a stronger influence. The regression equation, or the objective function model, is used to describe the impact of various factors on the process through a mathematical formula. Once this objective function model is accurately established based on experimental data, it can be used to predict the values of the objective function within the studied domain.

The second-order experimental matrix must possess the following basic properties [10]:

- Symmetry property: The sum of the vectors in each column x_j ($j \in \{1, 2, \ldots, k\}$, excluding the column corresponding to the intercept term ($j = 0$) must be equal to 0:

$$\Sigma(x_j)_u = 0 \tag{2}$$

- Orthogonality property: The sum of the products of any two vectors in the same row must be equal to 0:

$$\Sigma(x_i x_j)_u = 0 \quad (i, j = 1, 2, \ldots, k; j \neq i) \tag{3}$$

- Equidistance property: The sum of the squared vectors in each column must be equal to n:

$$\Sigma(x_j)_u^2 = n \tag{4}$$

In statistical experimental design, the number of experiments N increases with the number of factors k ($N = n^k$), and the number of experiments grows rapidly with the number of coefficients to be determined as k increases.

The total number of experiments in the design is calculated as follows:

$$\begin{cases} N = 2^k + 2k + n_0, k < 5 \\ N = 2^{k-p} + 2k + n_0, k > 5 \end{cases} \tag{5}$$

Where 2^k is the number of experiments in the factorial design TYT 2^k; n_0 is the number of experiments at the center point (n_0=1); $2k$ is the number of additional experiments. For a central composite design with three influencing factors (k = 3), we have:

$$N = 2^k + 2k + n_0 = 15 \tag{6}$$

2.3. Experimental procedure

A total of 17 experiments were carried out according to a central composite experimental design. This includes a full factorial design with 3 factors at two levels to estimate experimental error and check reproducibility. Using the levels in table 3, the experimental matrix was constructed as shown in table 4, which lists the actual experimental conditions for each run. It should be noted that other factors, such as heating rate and pressing rate, can also affect the transmittance by influencing internal stress and microstructure. In this study, these parameters were kept constant to focus on the effects of temperature, time, and pressure. Further studies will consider their influence in more detail.

Table 3. *Experimental factor levels and actual values for the design.*

Factor (unit)	- ω	-1	0	1	ω
x_1-T (°C)	730	750	850	950	970
x_2-t (minutes)	27	30	45	60	63
x_3-P (MPa)	190	200	250	300	310

Table 4. *Experiment parameters.*

Run	x_1-T (°C)	x_2-t (minutes)	x_3-P (MPa)
1	750	30	200
2	950	30	200
3	750	60	200
4	950	60	200
5	750	30	300
6	950	30	300
7	750	60	300
8	950	60	300
9	970	45	250
10	730	45	250
11	850	63	250
12	850	27	250
13	850	45	310
14	850	45	190
15	850	45	250
16	850	45	250
17	850	45	250

- Hot pressing: The sample was heated to the desired temperature, then the pressure was gradually increased to the setpoint and maintained for the required time duration (isothermal holding under pressure). This process consolidates the nanopowder into a dense optical ceramic.

- Product removal: After pressing, the assembly was allowed to cool down in air for a period of time before disassembling the mold.

- Test specimens: The test specimen of CaF_2 ceramic was prepared with the following geometry: outer diameter Ø67.5 mm, curvature radii $R_1 = 33$ mm and $R_2 = 37.5$ mm, and thickness 5.5 mm. We have added these details to the manuscript.

- Transmittance measurement: The optical transmittance of the CaF_2 ceramic samples, fabricated through the hot-pressing process, was evaluated using a Nicolet Summit PRO FTIR Spectrometer at the Measurement Center (Institute of Technology – VDI). Prior to measurement, the samples were carefully prepared to meet the requirements for accurate spectroscopic analysis. Specifically, both surfaces of each polycrystalline ceramic sample were machined to ensure parallelism within a tolerance of ≤ 0.01 mm, and the surface roughness was controlled to approximately Rz = 0.05 µm. The overall dimensions of the samples were tailored to fit precisely within the spectrometer's sample chamber.

3. RESULTS AND DISCUSSION

The transmittance results for all 17 experiments are presented in table 5. The table lists each run with its corresponding temperature, time, pressure, and the measured transmittance at 4.5 µm.

Table 5. *Experimental results – Transmittance at 4.5 μm.*

Run	x_1-T (°C)	x_2-t (minutes)	x_3-P (MPa)	Transmittance at 4,5 µm(%)
1	750	30	200	55
2	950	30	200	63
3	750	60	200	64
4	950	60	200	67
5	750	30	300	60
6	950	30	300	65
7	750	60	300	70
8	950	60	300	59
9	970	45	250	72
10	730	45	250	61
11	850	63	250	76
12	850	27	250	72
13	850	45	310	75
14	850	45	190	65
15	850	45	250	81
16	850	45	250	80
17	850	45	250	82

The second-order regression equation with three factors is as follows:

$$\begin{aligned} f &= b_0x_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1x_2 \\ &+ b_{13}x_1x_3 + b_{23}x_2x_3 + b_{11}(x_1^2 - \beta) + b_{22}(x_2^2 - \beta) + b_{33}(x_3^2 - \beta) \end{aligned} \tag{7}$$

with $\beta = 0,73$.

- Step 1: Calculate the regression coefficients of the second-order orthogonal design equation: The regression coefficients of the second-order regression equation are calculated using the following formula [9]:

$$b_i = \frac{\sum_{u=1}^{N} x_{iu}y_u}{\sum_{u=1}^{N} x_{iu}^2}; b_{ij} = \frac{\sum_{u=1}^{N} x_{iu}x_{ju}y_u}{\sum_{u=1}^{N} \left(x_{iu}x_{ju}\right)^2}; b_{ii} = \frac{\sum_{u=1}^{N} \left(x_{iu}^2 - \beta\right)y_u}{\sum_{u=1}^{N} \left(x_{iu}^2 - \beta\right)^2} \tag{8}$$

Where y_u represents the observed response value (objective function) in the u^{th} experiment. It is the output measured during the experiment corresponding to a specific set of input factor levels.

Table 6. *Estimated regression coefficients.*

b_0	b_1	b_2	$b3$	b_{12}	b_{13}	b_{23}	b_{11}	b_{22}	b_{33}
67.00	1.68	2.00	1.57	-2.63	-2.13	-1.13	-7.87	-2.79	-5.50

- Step 2: First, we calculate the reproducibility variance from the three replicate experiments at the center point and consider it as the common error for all experiments. The Student's t_α distribution is used to assess the significance of the regression coefficients [9].

$$t_i > t_\alpha(\alpha, f) \tag{9}$$

Where:

- t_i, t_{ij}, t_{ii} are the Student's t-statistics calculated for each regression coefficient, respectively. They are used to test whether each coefficient is significantly different from zero. The formula is:

$$t_i = \frac{|b_i|}{S_{b_i}}; t_{ij} = \frac{|b_{ij}|}{S_{b_{ij}}}; t_{ii} = \frac{|b_{ii}|}{S_{b_{ii}}} \tag{10}$$

with S_{bi} is the standard deviation (square root of the variance) of the coefficient b_i, and calculated by:

$$S_{b_i}^2 = \frac{S_0^2}{\sum (x_{iu})^2}; S_{b_{ii}}^2 = \frac{S_0^2}{\sum (x_{iu}^2 - \beta)^2}; S_{b_{ij}}^2 = \frac{S_0^2}{\sum (x_{iu} x_{ju})^2}; \tag{11}$$

Here, S_0 is the residual standard deviation, or the standard deviation of reproducibility errors obtained from the repeated measurements at the center point. It is used as the basis for estimating the variance of all regression coefficients.

- $t_\alpha(\alpha, f)$ is the critical value from the Student's t-distribution at a significance level α (typically 0.05) and $f = (N_0 - 1)$ is the degrees of freedom (where N_0 is the number of replications at the center point). It serves as the threshold to determine statistical significance: If $t_i > t_\alpha(\alpha, f)$ the corresponding coefficient is considered statistically significant.

Table 7. *Variance of the regression coefficients.*

$S_{b_0}^2$	$S_{b_1}^2$	$S_{b_2}^2$	$S_{b_3}^2$	$S_{b_{12}}^2$	$S_{b_{13}}^2$	$S_{b_{23}}^2$	$S_{b_{11}}^2$	$S_{b_{22}}^2$	$S_{b_{33}}^2$
0.07	0.09	0.09	0.09	0.13	0.13	0.13	0.23	0.23	0.23

Determine the statistical tests for the regression equation t_i:

Table 8. *t-test values for regression coefficients.*

t_{b_0}	t_{b_1}	t_{b_2}	t_{b_3}	$t_{b_{12}}$	$t_{b_{13}}$	$t_{b_{23}}$	$t_{b_{11}}$	$t_{b_{22}}$	$t_{b_{33}}$
259.75	5.55	6.61	5.18	7.42	6.01	3.18	16.78	6.18	11.83

After comparing the statistical test values with the critical value of Student's t distribution t_α (0,05;2) = 4,3, we obtain the regression coefficients as shown in the following table:

Table 9. *Regression coefficients after significance testing.*

b_0	b_1	b_2	b_3	b_{12}	b_{13}	b_{23}	b_{11}	b_{22}	b_{33}
67.00	1.68	2.00	1.57	- 2.63	- 2.13	0	- 7.87	- 2.79	- 5.50

Thus, we eliminate the coefficient b_{23}. After removing the non-significant coefficients, the resulting regression equation is as follows:

$$\begin{aligned} f &= 67 + 1.68x_1 + 2x_2 + 1.57x_3 + 2.63x_1x_2 - 2.13x_1x_3 - 7.87(x_1^2 - 0.73) \\ &-2.79(x_2^2 - 0.73) - 5.5(x_3^2 - 0.73) \end{aligned} \tag{12}$$

Take $x_1 = \frac{T - 850}{100}$; $x_2 = \frac{t - 45}{15}$; $x_3 = \frac{P - 250}{50}$ into (12) equation:

$$\begin{aligned} f &= -838.14 + 1.54T + 2.74t + 1.494P - 0.00175Tt - 0.000426TP \\ &- 0.0008T^2 - 0.0124t^2 - 0.0022P^2 \end{aligned} \tag{13}$$

- Step 3: Check the compatibility of the regression equation. The Fisher criterion is used to evaluate the adequacy of the regression equation.

$$F < F_\alpha(\alpha, f_1, f_2) \tag{14}$$

Where: $F = \frac{S_{du}^2}{S_0^2}$ với $S_{du}^2 = \frac{1}{N - L} \sum_{i=1}^{1} (Y_i - y_i)^2$; N is the number of experiments; L is the number of significant coefficients in the regression equation; $f_1 = N - L$; $f_2 = N_0 - 1$. If $F < F_\alpha(\alpha, f_1, f_2)$ then the equation is considered adequate.

From table 10, we see that $F < F_\alpha$, so the model is considered adequate. Therefore, the regression equation written in form (14) is reliable.

- Determination of optimal parameters: We used Maple 2022 software (Waterloo Maple Inc., Waterloo, Ontario, Canada) to determine the optimal parameters of the equation. The results show that the sample achieves the highest transmittance of 79.27% with the following optimal parameters: Hot-pressing temperature: T = 853 °C; Hot-pressing time: t = 50.2 minutes; Hot-pressing pressure: P = 256.8 MPa.

***Table 10.** Perform the regression equation test using the Fisher criterion.*

Residual variance S^2_{re}	15.49
f_1	6
f_2	2
Fisher criterion	
F	15.49
F_α	19.33

- Experimental validation: We verified the results through experiments using the following parameters: Hot-pressing temperature: 855 °C; Hot-pressing time: 50 minutes; Hot-pressing pressure: 255 MPa. The measured transmittance after conducting the experiment was as follows:

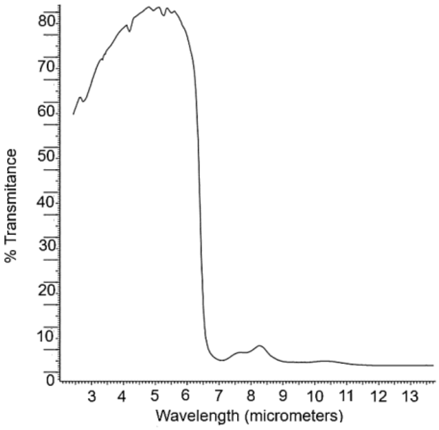

***Figure 2.** Measured transmittance results for the sample.*

The transmittance of the sample reached 82% at 4.5 µm (with a deviation of 3.3% from the predicted value), which confirms that the research method we are using is correct.

4. CONCLUSIONS

In this study, a comprehensive investigation of CaF_2 optical ceramic material and its hot-pressing process has been conducted. The structural characteristics of the material and its optical transmission mechanism were examined, along with their influence on the transmittance of CaF_2 optical ceramics. The research identified that among various fabrication factors, the hot-pressing temperature, pressure, and time are the three primary factors affecting the sintering process and the resultant optical transmittance of CaF_2 ceramics. A quadratic regression model relating these factors to the transmittance was developed and experimentally validated. Using this model, the optimal set of hot-pressing parameters for maximizing the transmittance was determined (approximately 853 °C, 50 min, 257 MPa in this study), and the model-predicted improvement in transmittance was confirmed by a verification experiment. It is noted that hot-pressing is a complex process. Apart from selecting appropriate temperature, pressure, and time, other factors such as the heating and pressurization rates, the atmosphere of pressing, and the mold material can also influence the quality of the optical ceramic product. These factors were beyond the scope of the current study and present opportunities for future research. Further analysis on those aspects could provide additional insights and lead to improved processes for manufacturing high-quality optical ceramics under various production conditions.

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TÓM TẮT

Ảnh hưởng của các tham số công nghệ đến quá trình ép nóng gốm quang học CaF_2

Bài báo sử dụng phương pháp quy hoạch thực nghiệm nghiên cứu ảnh hưởng của các thông số công nghệ đến quá trình ép nóng của gốm quang học CaF_2. Mô hình quy hoạch thực nghiệm được đưa vào để xây dựng các phương trình tương quan giữa các thông số công nghệ như nhiệt độ, thời gian, và áp suất đến chỉ số chất lượng của độ truyền qua của gốm quang học. Từ đó, các tác giả xác định được bộ thông số công nghệ ép nóng tối ưu cho quá trình ép nóng gốm quang học CaF_2. Kết quả nghiên cứu của bài báo là tiền đề quan trọng để mở ra nhiều hướng nghiên cứu khác nhau cho quá trình công nghệ ép nóng các loại gốm quang học khác nhau sử dụng trong các thiết bị khí tài quân sự.

Từ khoá: Quy hoạch thực nghiệm; Gốm quang học CaF_2; Ép nóng; độ truyền qua; Khí tài quang.