



HAL
open science

Determination Of The Refractive Index Of Optical Ceramics By Using Transmission Data In The Mid-Wave Infrared Region

Mai van Huy, Duong Chi Dung, Le Hoang Hai, Le Duy Hoan, Nguyen Tuan
Hieu, Olivier Schneegans

► **To cite this version:**

Mai van Huy, Duong Chi Dung, Le Hoang Hai, Le Duy Hoan, Nguyen Tuan Hieu, et al.. Determination Of The Refractive Index Of Optical Ceramics By Using Transmission Data In The Mid-Wave Infrared Region. Journal of Science and Technique, 2020, 10.56651/lqdtu.jst.v15.n04.7 . hal-03090366

HAL Id: hal-03090366

<https://centralesupelec.hal.science/hal-03090366>

Submitted on 5 Feb 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

DETERMINATION OF THE REFRACTIVE INDEX OF OPTICAL CERAMICS BY USING TRANSMISSION DATA IN THE MID-WAVE INFRARED REGION

Van Huy Mai^{1*}, Chi Dung Duong¹, Hoang Hai Le¹, Duy Hoan Le², Tuan Hieu Nguyen², Olivier Schneegans³

¹ Department of Optical Devices, Le Quy Don Technical University, 236 Hoang Quoc Viet, Hanoi

² Institute of Technology, Viet Nam of Defense Industry, Duc Thang, Bac Tu Liem, Hanoi

³ Laboratoire Génie Électrique et Électronique de Paris (GeePs) - CNRS – CentraleSupélec, Sorbonne Université and Université Paris-Saclay – 91190 Gif-sur-Yvette, France

*E-mail: vanhuy.mai@mta.edu.vn

Abstract:

Optical ceramics are emerging materials for optical devices operating in the Mid-Wave Infrared (MWIR) spectral range. Thus determination of their refractive index is of huge importance but is still a challenging issue. In this work, a model is delivered, which highlights a simple mathematic relationship between the refractive index of an optical material and its transmittance spectrum measured by Fourier Transform Infrared spectroscopy. Such model allows determining the refractive index of ceramics fabricated by hot-pressing of MgF₂ nanoparticles. The reliability of this method has been verified on a silicon wafer taken as a reference (< 0.5%, discrepancy with data from literature on Si). We believe that our approach can be applied to a variety of promising novel optical materials being developed.

Keyword: refractive index, optical ceramic, FTIR (Fourier Transform Infrared Spectroscopy)

1. Introduction

In the design and fabrication of an optical system, the refractive indexes of the constitutive materials are important parameters to determine. Towards such a characterization, many methods have been proposed in the past, ranging from classical to newer techniques [1-4]. These can be grouped into two main categories: prism methods and spectral methods. Regarding the prism methods, the deflection of a beam entering and leaving a prism is measured. The advantage of these methods (e.g. measurement of deviation, or minimum deviation angle, by refractometry, for which a variety of different techniques have been developed historically [1]) is that an accurate value of refractive index can be obtained at a fixed wavelength. This implies the use of an expensive refractometer and the fabrication of a prism with high accuracy [1]. Unfortunately, these methods appear not suitable to measure the refractive index over a wide range of wavelengths. Hence spectral methods using

transmission and reflection data have increasingly been applied to determine the refractive index of materials with a good accuracy. In this case however, each time a new kind of materials is studied, special care is required to ensure that the proposed model is in adequation with the experimental data [5, 6].

Single-crystal magnesium fluoride (MgF_2) is a positive uniaxial crystal whose useful optical transmission is in the range of between 0.11 and $7.5\mu\text{m}$ [3]. An optical ceramic, composed of polycrystalline high density MgF_2 , is elaborated by a hot-pressing technique; in the MWIR, its high transmittance interval lies in the range 2.5 to $6\mu\text{m}$ [7]. This material has been widely used in the field of optics, especially for missiles domes. By traditional methods, the refractive index of this material is difficult to determine in the MWIR. In this report, we first present the calculation and experiments carried out to determine the refractive index of a Si substrate by using Fourier Transform Infrared Spectroscopy (FTIR). Our results were in good agreement ($<0.5\%$ discrepancy) with published results in the 3-5 μm spectral range [8], hence we then applied this method to determine the refractive index of an optical ceramic, using parallel plate samples.

2. Methods and Experiments

Mathematical model for calculation of refractive index

Towards calculation of the refractive index, we first assume that a flat parallel plate sample has been polished on both sides (the surface roughness may thus be neglected), and that the sample is surrounded by ambient air (conditions to measure the transmission data), as illustrated in figure 1. The light beam reaches the sample at normal incidence, the sample thickness is such as both interference and absorption can be neglected.

For each interface (air/substrate and substrate/air, respectively), we denote the transmittance and reflectance: T_1 , R_1 and T_2 , R_2 respectively. For a light (of intensity I_0) coming from the left (figure 1), we can calculate the intensity at each interface by using the following equations [9]:

$$I_1 = T_1 \cdot I_0 \quad (1)$$

$$I_2 = T_2 \cdot I_1 = T_1 \cdot T_2 \cdot I_0 \quad (2)$$

$$I_3 = R_2 \cdot I_1 = R_2 \cdot T_1 \cdot I_0 \quad (3)$$

$$I_4 = R_1 \cdot I_3 = R_1 \cdot R_2 \cdot T_1 \cdot I_0 \quad (4)$$

$$I_5 = T_2 \cdot I_4 = T_1 \cdot T_2 \cdot R_1 \cdot R_2 \cdot I_0 \quad (5)$$

$$I_6 = R_2 \cdot I_4 = R_1 R_2^2 \cdot T_1 \cdot I_0 \quad (6)$$

$$I_7 = R_1 \cdot I_6 = R_1^2 \cdot R_2^2 \cdot T_1 \cdot I_0 \quad (7)$$

$$I_8 = T_2 \cdot I_7 = T_1 \cdot T_2 \cdot (R_1 \cdot R_2)^2 \cdot I_0 \quad (8)$$

...

$$I_{tot} = I_2 + I_5 + I_8 + \dots \quad (9)$$

$$I_{tot} = T_1 \cdot T_2 \cdot \left[I + R_1 \cdot R_2 + (R_1 \cdot R_2)^2 + (R_1 \cdot R_2)^3 \dots \right] \cdot I_0 \quad (10)$$

Since R_1 and R_2 are both inferior to 1, we can re-calculate:

$$I + R_1 \cdot R_2 + (R_1 \cdot R_2)^2 + (R_1 \cdot R_2)^3 \dots = \frac{I}{1 - R_1 \cdot R_2} \quad (11)$$

Therefore, expression (10) becomes:

$$I_{tot} = T \cdot I_0 = \left(\frac{T_1 \cdot T_2}{1 - R_1 \cdot R_2} \right) \cdot I_0 \quad (12)$$

where the total transmittance T is:

$$T = \frac{T_1 \cdot T_2}{1 - R_1 \cdot R_2} \quad (13)$$

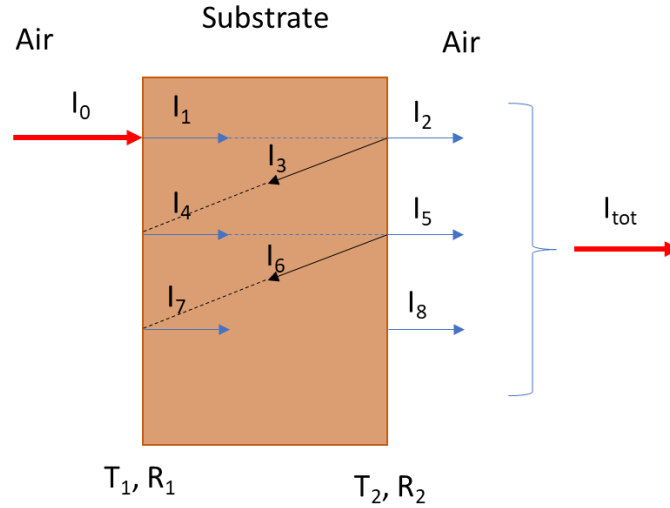


Figure 1: Schematics of the light transmission through a parallel plate substrate in air. The light (intensity I_0) comes from the left. The resulting total transmitted light (I_{tot}) depends on T_1, R_1, T_2 and R_2

If we assume that both sides of the sample have undergone the same polishing, then both interfaces (air/substrate and substrate/air) are completely symmetric, hence $T_1 = T_2$ and $R_1 = R_2$. Besides, for a transparent sample (absorbance $A = 0$), we have $T_1 = 1 - R_1$. Thus the total transmittance T of the sample can be retrieved from (13):

$$T = \frac{(1-R_1)^2}{1-R_1^2} \quad (14)$$

where R_1 is the reflectance which comes from the Fresnel equations:

$$R_1 = \left(\frac{n-1}{n+1} \right)^2 \quad (15)$$

Finally, as the transmittance T is a function of wavelength λ , the refractive index $n(\lambda)$ is obtained from the transmittance $T(\lambda)$ using (14) and (15):

$$n(\lambda) = \frac{1 + \sqrt{1-T(\lambda)^2}}{T(\lambda)} \quad (16)$$

Fabrication of the IR optical ceramics

The experiments have been carried out on two kinds of samples:

- a Si wafer (diameter 1 inch, thickness 2mm, polished on both sides), with the objective of comparing our results with published data. This kind of wafer is almost used in the field of optical thin film coating.

- two optical ceramic parallel plate samples which were fabricated at the Institute of Technology, Vietnam of Defense Industry. The samples were fabricated by hot pressing of MgF_2 nano particles (size 70-90nm), in an annealing furnace at a pressure of 10^{-2} bar. The optical ceramic parallel plate samples M1 and M2 have been polished on both sides (resulting roughness $< 0.05\mu\text{m}$). Sample M1 (thickness 1.032mm) was annealed at a temperature of 640.9°C during 33 minutes at a pressure of 231.4MPa, and sample M2 (thickness 1.842mm) has been annealed at a temperature of 651°C during 35 minutes at a pressure of 240.5MPa. The mass density of samples M1 and M2 are 3.17 and 3.18g/cm^3 respectively. The transmittance of the samples was measured in the MWIR region by using a FTIR spectrometer “Nicolet Summit Pro” of Thermo Scientific.



Figure 2: Optical photos of the optical ceramic samples: the thickness of M1 (left) is 1.032mm, the thickness of M2 (right) is 1.842mm

Results and discussion

In order to estimate the mathematical model derived in section 2, we first measured the transmission of the Si wafer (shown in figure 3). Then, by using formula (16), the refractive index of the Si wafer has been determined (black curve in figure 4). Cauchy's dispersion formula is a well-known empirical relationship between the refractive index and wavelength for transparent materials [10-14], and has also been used to derive the refractive index of chalcogenide glasses in the NIR (near infrared) – MWIR spectral range [15]. Hence, we used in our case such a Cauchy law to fit (with an iterative Levenberg Marquardt algorithm) the refractive index data of the Si wafer (red curve in figure 4). The results obtained for Si are in good agreement ($< 0.5\%$ discrepancy) with the results of reference [8] (ten star points in figure 4) over the range $[3-5\mu\text{m}]$. Although the accuracy of our results maybe even better with higher resolution transmission data, they were already very promising, hence this method has been applied to study the optical ceramic materials, for which there are no references concerning measurements of refractive index.

For the optical ceramic samples, we observe that in the range $[3-5\mu\text{m}]$, the transmittance of sample M1 (figure 5) increases from about 91% to 96%, and that the transmittance of sample M2 (figure 6) increases from 84% to 91% as a result of different conditions of fabrication (including different mass densities).

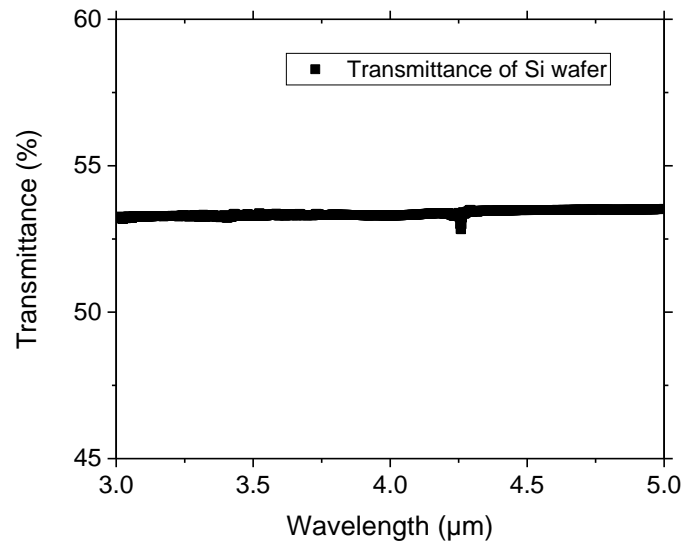


Figure 3. Transmittance spectrum of the Si wafer

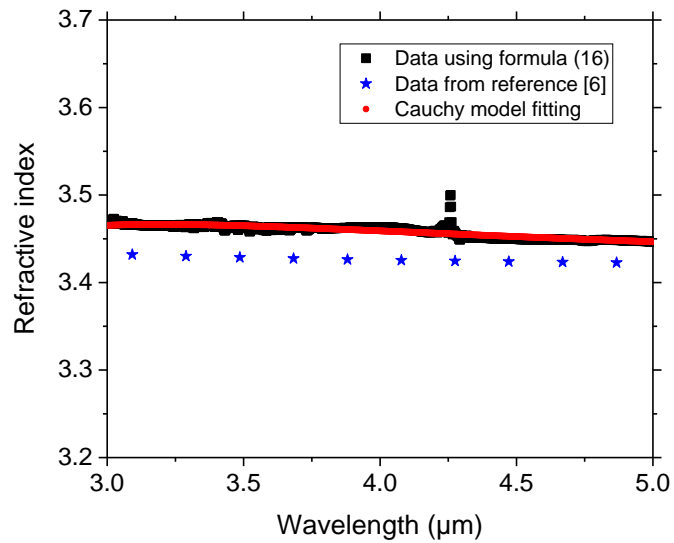


Figure 4. Refractive index of Si: square dots curve is calculated from transmittance data; circular dots curve is the Cauchy model fitting, star points are data from reference [8]

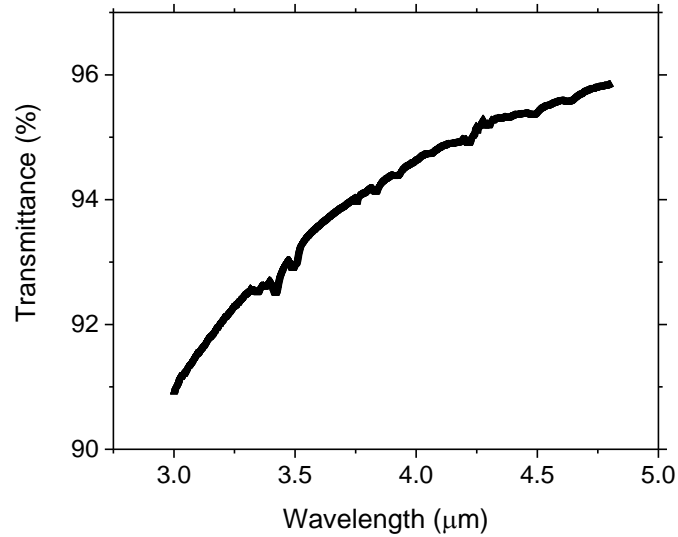


Figure 5: Transmittance spectrum of sample M1 (thickness 1.032mm)

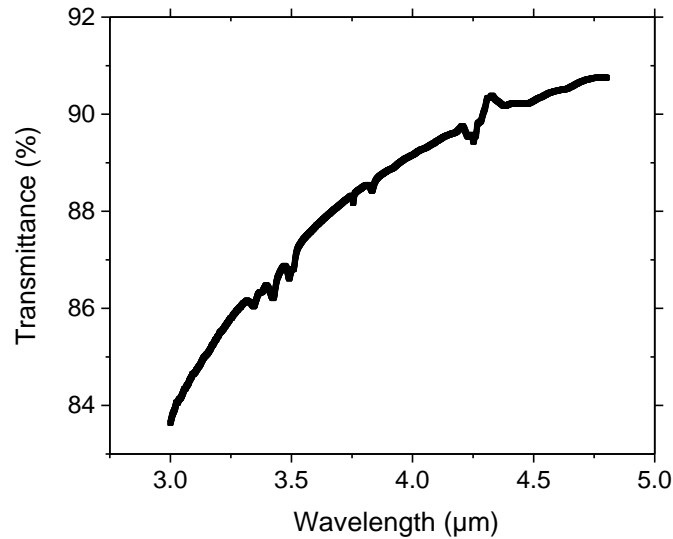


Figure 6: Transmittance spectrum of sample M2 (thickness 1.842mm)

By using formula (16), the refractive indexes of the optical ceramic samples can be obtained, as illustrated in figures 7 and 8. By looking at these curves, we can see that there are some little peaks (which may come from losses in the materials due to any reason). Apart from these isolated peaks, as the optical ceramic samples were fabricated from MgF_2

nanoparticles, the absorption may be very low in the MWIR region. From these results, we found that the refractive index is well fitted with such Cauchy model (figures 7 and 8, line-curves), and we see the high influence of fabrication conditions on the refractive index of the ceramic materials.

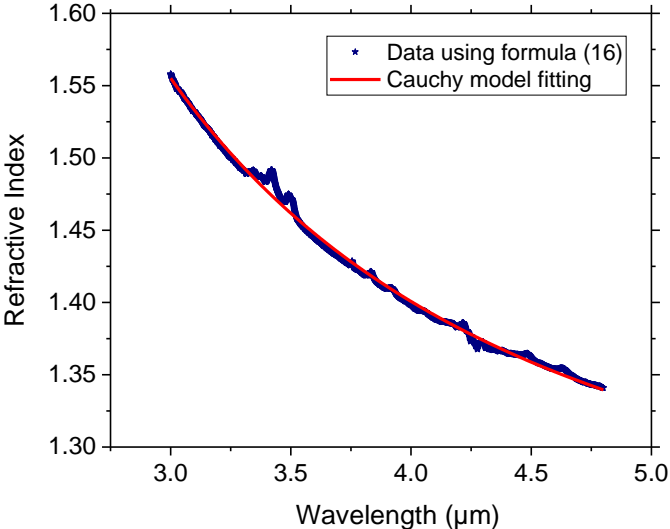


Figure 7: Black curve: refractive index of sample M1 (thickness 1.032mm), obtained from transmission data (figure 5) using (16). Red curve: fit with a Cauchy-law: $a + b/\lambda^2 + c/\lambda^4$

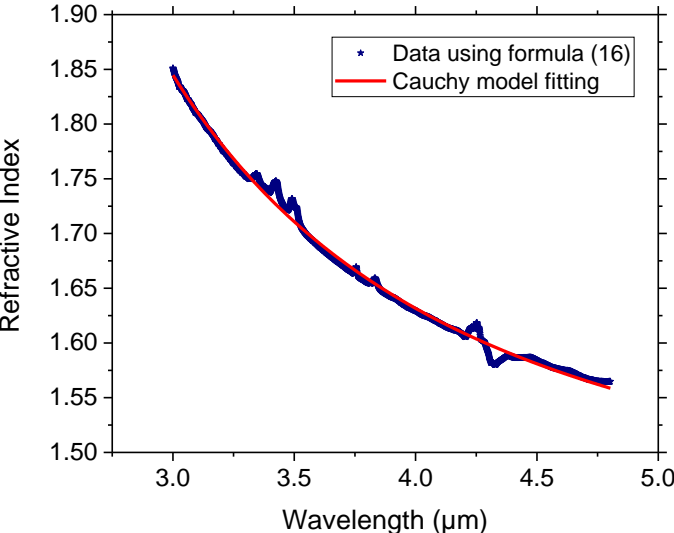


Figure 8: Black curve: refractive index of sample M2 (thickness 1.842mm), obtained from transmission data (figure 6) using (16). Red curve: fit with a Cauchy law: $a + b/\lambda^2 + c/\lambda^4$

Table 1 below summarizes the parameters found by using a Cauchy formula which fits well the refractive index evolution of samples M1 and M2 over a wide range of refractive index ([1.35-1.55] for M1 and [1.55 – 1.85] for M2) with a high accuracy.

Cauchy Dispersion Formula	$a + b/\lambda^2 + c/\lambda^4$	
Parameter	Sample M1	Sample M2
a	1.19945 ± 0.00116	1.41611 ± 0.00168
b	3.24547 ± 0.03199	2.92247 ± 0.04639
c	-0.39791 ± 0.2089	8.423 ± 0.30359

Table 1: Parameters of the Cauchy law, obtained by fitting the refractive index curves calculated from transmittance data (figures 7 and 8).

Conclusion

In this report, a simple method has been presented, in order to determine the refractive indexes of a Si wafer and two optical ceramic plates in the MWIR region, by use of transmission data. It has been verified that the refractive index evolution $n(\lambda)$ of the transparent materials are well fitted by a Cauchy dispersion formula. Based on the fact that this method is simple and does not require an expensive device for measurements, it may provide a tool in the development of fabrication technology of optical ceramics and in the determination the refractive index of novel optical materials.

References

1. <http://www.refractometer.pl/Abbe-refractometer>.
2. Gao, L., F. Lemarchand, and M. Lequime, *Refractive index determination of SiO₂ layer in the UV/Vis/NIR range: spectrophotometric reverse engineering on single and bi-layer designs*. 2013. Vol. 8. 2013.
3. Dodge, M.J., *Refractive properties of magnesium fluoride*. Applied Optics, 1984. **23**(12): p. 1980-1985.
4. Fang, Y., et al., (INVITED) *Methods for determining the refractive indices and thermo-optic coefficients of chalcogenide glasses at MIR wavelengths*. Optical Materials: X, 2019. **2**: p. 100030.
5. Gerstenlauer, D., et al., *Determination of the refractive index of TiO₂-Y₂O₃ ceramic composites*. Journal of Quantitative Spectroscopy and Radiative Transfer, 2014. **135**: p. 44-49.
6. Mehling H, KuhnJ, ManaraJ, BrandtR, FrickeJ. *Different methods for determining the refractive index of SiO₂ glass in the range of 1.4–50μm*. High Temp High Press 2001; **33**: 35–42.
7. Chang, C.S., M.H. Hon, and S.J. Yang, *The optical properties of hot-pressed magnesium fluoride and single-crystal magnesium fluoride in the 0.1 to 9.0 μm range*. Journal of Materials Science, 1991. **26**(6): p. 1627-1630.
8. Chandler-Horowitz, D. and P.M. Amirtharaj, *High-accuracy, midinfrared (450cm⁻¹ ≤ ω ≤ 4000cm⁻¹) refractive index values of silicon*. 2005. **97**(12): p. 123526.
9. Olaf Stenzel, Chapter 7 of "The Physics of Thin Film Optical Spectra, An Introduction" (Second Edition), Springer Series in Surface Sciences, 2016.
10. Liu, J.-m., *Photonic Devices*. 2005, Cambridge: Cambridge University Press.
11. Jr., G.E.J. and F.A. Modine, *Parameterization of the optical functions of amorphous materials in the interband region*. 1996. **69**(3): p. 371-373.
12. F.A. Jenkins, H.E. White, *Fundamentals of Optics*, Tata McGraw-Hill Education, 1937.
13. H.G. Dantanarayana, *Application of TLM for Optical Microresonators*, PhD thesis University of Nottingham, UK, 2012.
14. https://en.wikipedia.org/wiki/Cauchy%27s_equation.
15. Nĕmec, P., et al., *Optical properties of (GeSe₂)_{100-x}(Sb₂Se₃)_x glasses in near- and middle-infrared spectral regions*. Materials Research Bulletin, 2014. **51**: p. 176-179.

XÁC ĐỊNH CHIẾT SUẤT CỦA VẬT LIỆU GỐM QUANG HỌC TỬ PHỔ TRUYỀN QUA TRONG VÙNG HỒNG NGOẠI

Tóm tắt:

Gốm quang học là vật liệu mang tính cấp thiết trong các thiết bị quang học làm việc trong vùng hồng ngoại. Do đó, việc xác định chiết suất của gốm quang học là một nhiệm vụ quan trọng nhưng vẫn là một nhiệm vụ khó khăn. Trong bài báo này, chúng tôi dẫn chứng một mối quan hệ toán học đơn giản giữa chiết suất và hệ số truyền qua (FTIR), điều này cho phép xác định được chiết suất của gốm quang học được chế tạo bằng phương pháp ép nóng hạt nano MgF₂. Độ tin cậy của phương pháp đã được kiểm nghiệm trên đế Si và so sánh với các kết quả công bố với độ sai lệch dưới 0.5%. Chúng tôi tin tưởng rằng, phương pháp này có thể áp dụng để phát triển các vật liệu quang học mới.

Từ khóa: chiết suất, gốm quang học, phổ hồng ngoại truyền qua fourier.