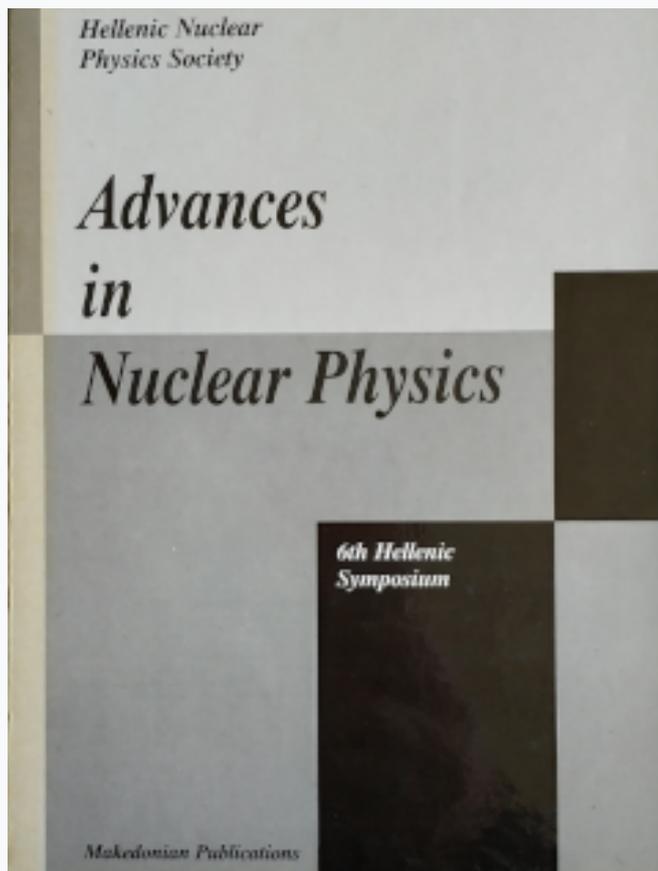


## HNPS Advances in Nuclear Physics

Vol 6 (1995)

HNPS1995



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doi: [10.12681/hnps.2935](https://doi.org/10.12681/hnps.2935)

#### To cite this article:

Aslanoglou, X., Assimakopoulos, P. A., Trapalis, C., Kordas, G., Karakassides, M. A., & Pilakouta, M. (2020). Heavy Ion RBS Characterization of Multilayer Coatings Deposited on Glass Through the Sol-Gel Technique. *HNPS Advances in Nuclear Physics*, 6, 279–287. <https://doi.org/10.12681/hnps.2935>

# Heavy Ion RBS Characterization of Multilayer Coatings Deposited on Glass Through the Sol-Gel Technique.

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## Abstract

Multilayer reflecting thin films of the systems  $\text{TiO}_2$ - $\text{SiO}_2$  and  $\text{ZrO}_2$ - $\text{SiO}_2$  were deposited on glass surface using the sol-gel technique. A  $^{12}\text{C}$  beam was utilized in RBS analysis to investigate the inner structure of these multilayer stacks. No evidence for defects or diffusion between the layers were found. Optical Spectroscopy showed wavelength selectivity in the reflection of an 8-layer  $\text{TiO}_2$ - $\text{SiO}_2$  sample.

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## 1 Introduction

Rutherford Backscattering Spectrometry using heavy ion (HIRBS) beams provides some interesting differences in comparison to the conventional  $^4\text{He}$  spectroscopy. As has been pointed out by many authors in a number of comparative studies [1-4], although the energy resolution of surface barrier detectors deteriorates as the mass of the particles and the energy of the beam increases [5], the overall mass resolution of the target improves with the increase of the mass and the energy of the beam. This behaviour follows the difference in the behaviour of the Kinematic Factor in different reactions. A calculation of the dependence of the mass resolution on the target mass number for the case of a  $^4\text{He}$  beam and a  $^{12}\text{C}$  beam is shown in fig. 1. The calculation was done for incident energy 2 MeV and detector resolution (FWHM) 20 keV for  $^4\text{He}$ , while for the  $^{12}\text{C}$  case the incident energy was 10 MeV and the detector resolution was 80 keV. As it emerges from this comparison, the use of heavy ion beams for HIRBS provides superior mass resolution than the conventional low

energy  $^4\text{He}$  scattering. The depth resolution remains at a comparable level in most case because the greater energy deposited by the heavy ion competes with the lower detector resolution. However, the lowest detectable mass number is higher in HIRBS, because elements with mass number smaller, or around the mass number of the beam are invisible in backscattering.

In this work we have used a  $^{12}\text{C}$  beam at energy  $E=10$  MeV to investigate the structure of multi layer systems made of successive thin films of  $\text{TiO}_2\text{-SiO}_2$  or  $\text{ZrO}_2\text{-SiO}_2$  deposited on glass.

Thin films of metal oxides (dielectrics) on glass, either in a single layer or in multi layer coatings of different elements each, are known to modify the optical properties of the glass. A series of dielectric films with alternate high and low refractive indices can, under certain conditions, lead to selectivity in the reflection of only certain wavelengths [6,7]. In our case, the sequence of the low index of refraction of  $\text{SiO}_2$  and the high index of refraction of  $\text{ZrO}_2$  or  $\text{TiO}_2$  are tested for the production of a selective mirror. The optical properties of the systems were studied by Ultra Violet, Visible and Near Infra Red reflectance spectroscopy.

## 2 Experimental Procedure.

The sol-gel technique was used for the fabrication of the oxide films at the Institute of Materials Science at the National Center for Scientific Research (NCRS) "Demokritos", Athens, Greece. This technique is widely used and almost exclusively applied for the fabrication of transparent coatings for deposition on glass surface. The starting solutions for the preparation of the sol were  $\text{Si}(\text{C}_2\text{H}_5\text{O})_4$ ,  $\text{Ti}(\text{C}_2\text{H}_5\text{O})_4$  and  $\text{Zr}(\text{C}_2\text{H}_5\text{O})_4$  in ethanol, with  $\text{HNO}_3$  as a catalyst. More information about this technique can be found in ref. [8]. After the sol preparation, thin layers of metal oxides ( $\text{ZrO}_2$ ,  $\text{TiO}_2$  or  $\text{SiO}_2$ ) were deposited on flat glass plates of dimensions (60mm X 20mm X 1.5mm) using the dipping process. After the deposition of each new layer, the sample was dried at room temperature and then was heated in a furnace at  $500^\circ\text{C}$  for 20 min. This way, stacks of up to eight layers  $\text{TiO}_2\text{-SiO}_2$  were prepared and up to five layers  $\text{ZrO}_2\text{-SiO}_2$ . The RBS measurements were performed by utilizing a  $^{12}\text{C}$  beam of energy 10 MeV, from the 5.5 MV TN/11 TANDEM Van De Graaff accelerator of the Institute of Nuclear Physics, NCRS "Demokritos". The detection system consisted of a single Si surface barrier detector, placed at scattering angle  $\Theta=150^\circ$  with respect to the beam. For the analysis and the simulation of the spectra, a new code was developed, which is capable of handling any heavy ion beam. This code uses the Universal Energy Loss Coefficients by J.F. Ziegler [9] and the subroutine ZSTOP to calculate the normalized yield. The overall accuracy of the determination of the thickness

is 10%.

### 3 Results and Discussion.

#### 3.1 RBS Analysis

All the samples that were manufactured were subsequently characterized by RBS in search for defects in the internal structure. The spectra of two samples, a three layer  $ZrO_2$ - $SiO_2$ - $ZrO_2$  and a five layer  $ZrO_2$ - $SiO_2$ - $ZrO_2$ - $SiO_2$ - $ZrO_2$  are shown in fig. 2 and 3, together with the corresponding simulated spectra. The composition of the glass substrate was taken as 51% O, 43% Si and 6% Ca. As can be seen, this composition simulates well the substrate in all the cases. The plateau of  $^{16}O$  can not be seen in the spectrum, because Oxygen has a mass comparable to the Carbon beam and therefore is "invisible" in HIRBS. The RBS spectra indicate that the  $ZrO_2$  film is contaminated by an element of mass around 180 (with  $\Delta M=7$ , from fig. 1). A subsequent X-Ray Fluorescence (XRF) analysis showed that the  $ZrO_2$  employed for the manufacture of the samples was found to be contaminated by  $HfO_2$  at the level of 2%. In either case, the inner layers do not show any defects or significant diffusion effects due to the successive thermal treatment of the manufacturing process. The surface density of the  $ZrO_2$  and  $SiO_2$  films were found in either case  $27 \pm 1 \mu g/cm^2$  and  $19 \pm 1 \mu g/cm^2$ . The spectrum of an eight layer  $4(TiO_2-SiO_2)$  is shown in fig 4. In this sample, the layer neighbouring the glass substrate is  $SiO_2$ , which has a composition similar to the substrate and therefore can not be distinguished in the spectrum. Natural Titanium has six stable isotopes:  $^{46}Ti$  8%,  $^{47}Ti$  7.5%,  $^{48}Ti$  73.7%,  $^{49}Ti$  5.5% and  $^{50}Ti$  5.3%. The mass resolution with the reaction employed here, around the target mass number 50 is 2 (fig. 1). As a result, the minor isotopes can be distinguished from  $^{48}Ti$ , and they demonstrate themselves as tails around the major peak. To obtain a good simulation of the Ti peak, all six isotopes were included in the calculation as components of the layer. The result shows a good reproduction of the overlap between the Ti peaks in the simulation spectrum. A high degree of uniformity of the thicknesses of the layers was found for this sample, with the  $SiO_2$  layers having a surface density  $\rho_{Si} = 28 \pm 1 \mu g/cm^2$  and the  $TiO_2$  layers  $\rho_{Ti} = 13 \pm 1 \mu g/cm^2$ . Intermediate samples with fewer coatings showed the same behaviour.

#### 3.2 Reflectance Analysis

Reflectance spectra were obtained for both the Ti based and Zr based samples. Fig. 4 shows the reflectance spectra of the glass substrate covered with a

number of (TiO<sub>2</sub>-SiO<sub>2</sub>) films. The reflectance of the glass substrate (R=8.2%) increases according to the number of layers deposited and becomes maximum when 8 layers are deposited. The reflectance of this sample is R=85% in the wavelength region of 600-650 nm, thus becoming a selective mirror. In the case of the Zr based group, the transition from a single ZrO<sub>2</sub> layer to three and five ZrO<sub>2</sub> and SiO<sub>2</sub> stacks did not show any significant selectivity of the reflectance on a particular wavelength band.

#### 4. Conclusions.

In this work we used Heavy Ion RBS to study the structure of multi layer coatings deposited on glass substrate, by the sol-gel process. The employed 10 MeV <sup>12</sup>C beam was found to have good mass and depth resolution for analysis of thin films. Although the inner layers of the samples examined suffered multiple thermal treatments during fabrication, we found no evidence of defects or diffusion between the layers. The optical analysis showed that the eight layer (TiO<sub>2</sub>-SiO<sub>2</sub>) stack had an 85% reflectance in the wavelength region 600-650 nm.

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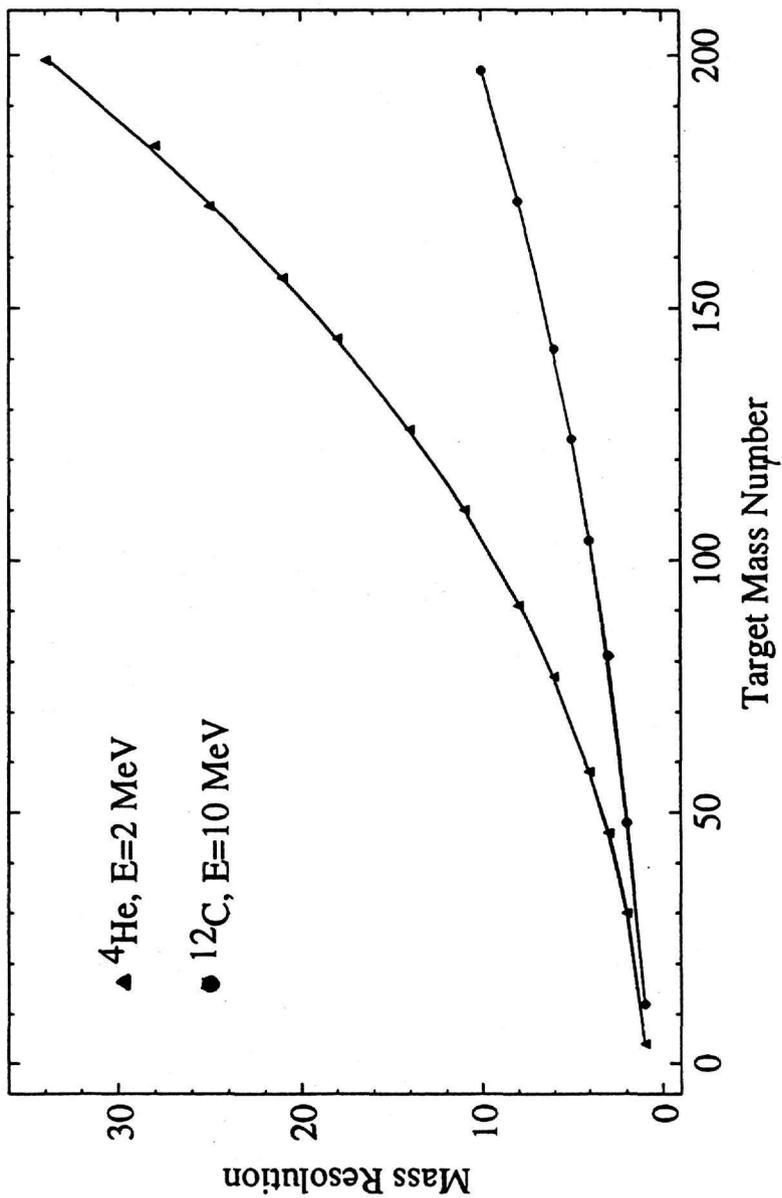


Fig. 1. Mass resolution versus target mass number for a 2 MeV  ${}^4\text{He}$  beam and detector resolution 20 keV and a  ${}^{12}\text{C}$  beam and detector resolution 80 keV.

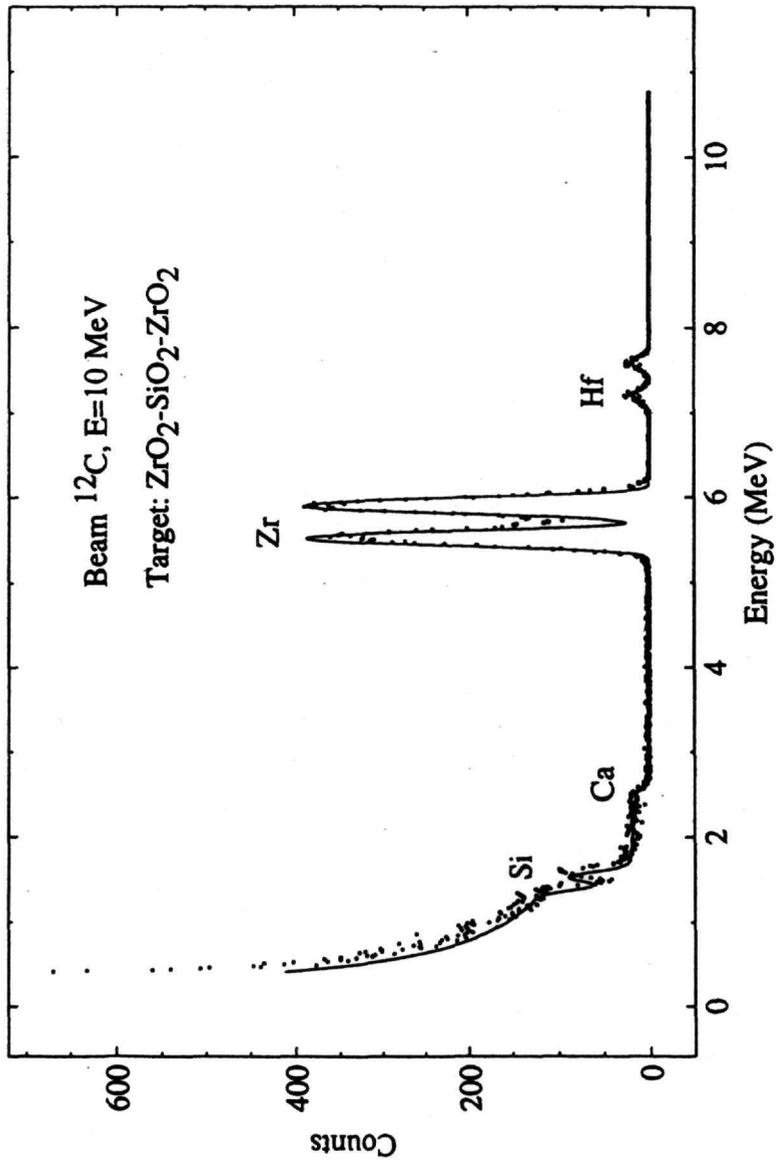


Fig. 2. RBS spectrum of a three layer  $\text{ZrO}_2\text{-SiO}_2\text{-ZrO}_2$  sample on glass substrate. The solid line represents the simulation spectrum.

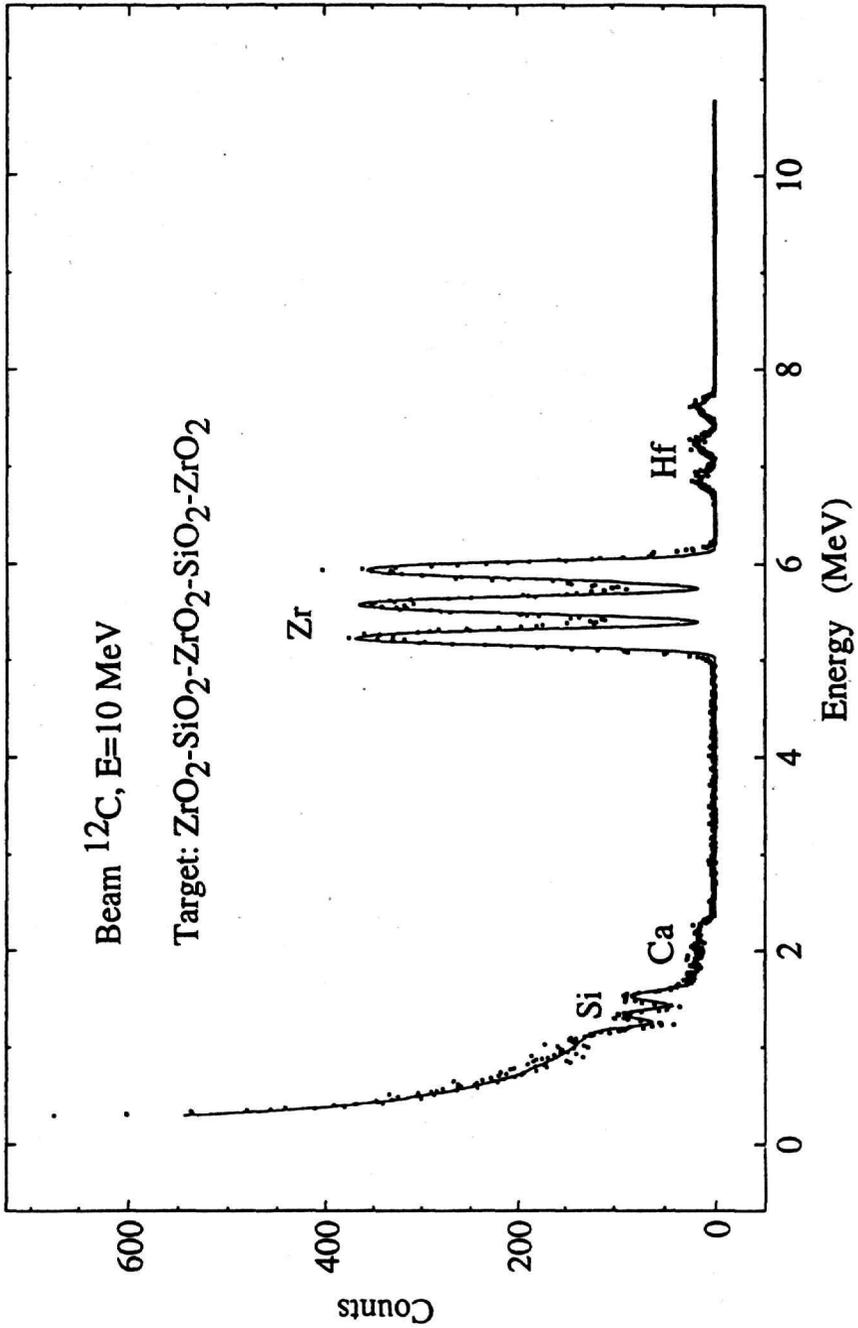


Fig. 3. RBS spectrum of a five layer ( $\text{ZrO}_2\text{-SiO}_2$ ) sample on glass substrate together with the corresponding simulation.

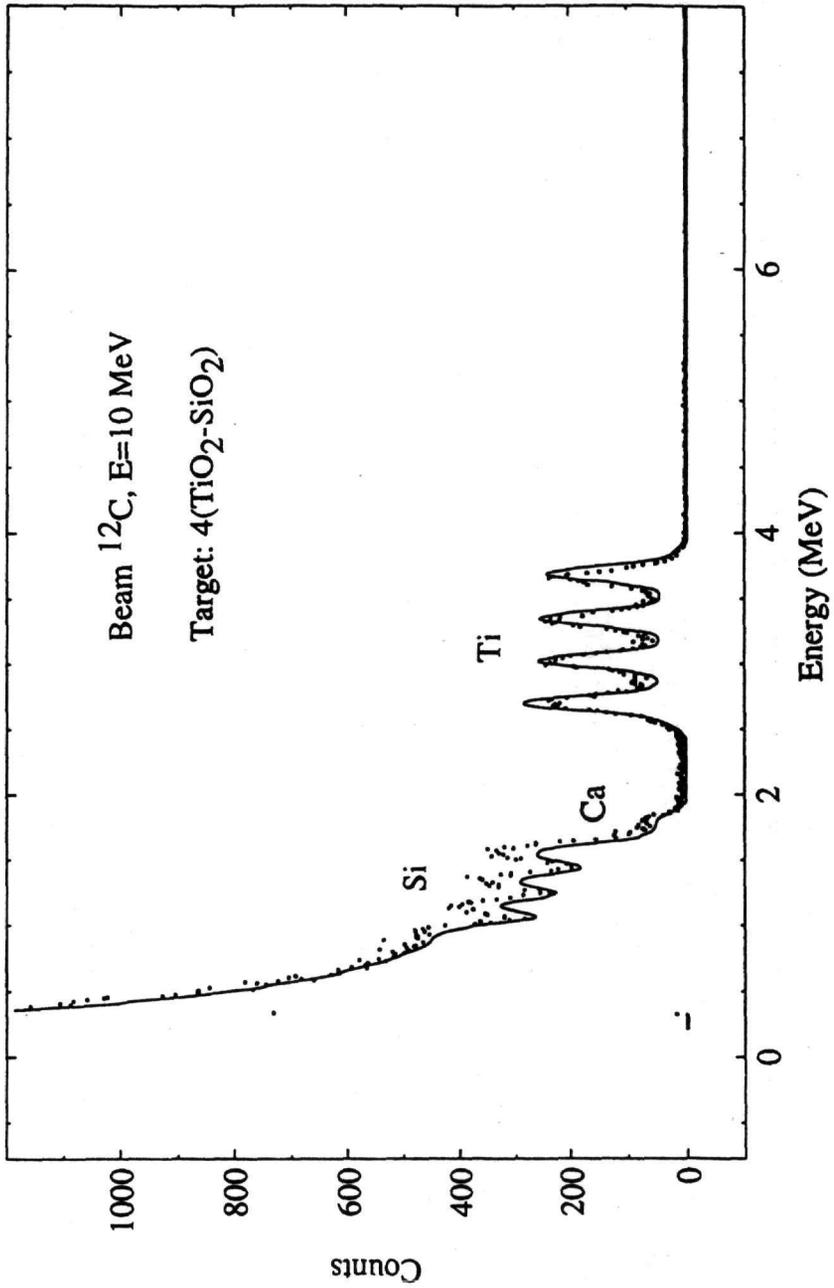


Fig. 4. RBS spectrum of an eight layer  $4(\text{TiO}_2\text{-SiO}_2)$  sample on glass substrate. The layer neighbouring to the glass is  $\text{SiO}_2$  and can not be distinguished.

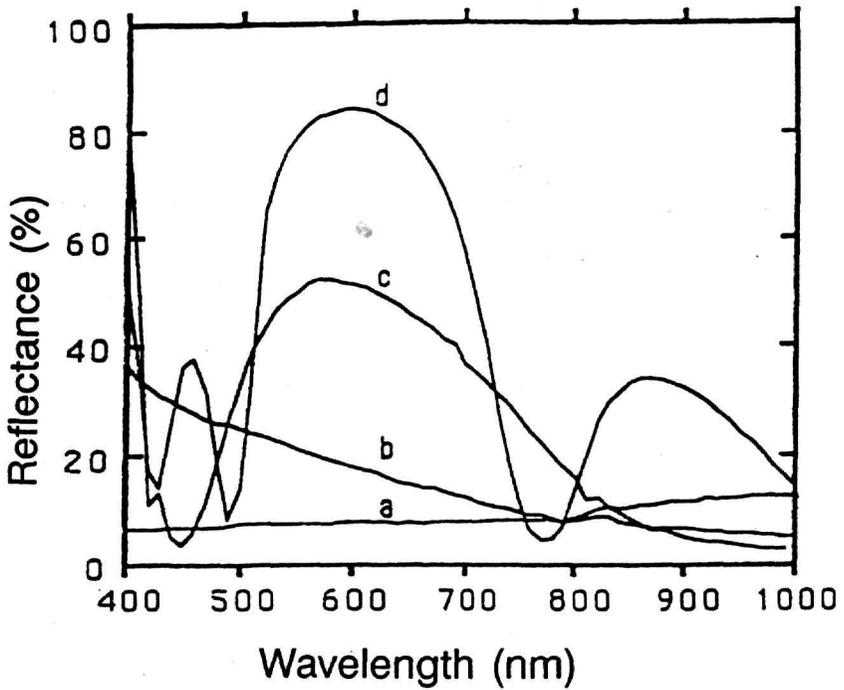


Fig. 5. Reflectance analysis for the eight layer  $4(\text{TiO}_2\text{-SiO}_2)$  sample shown in fig. 4. a) substrate b) two layer stack c) four layer stack d) eight layer stack.