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PII: S0169-4332(18)31794-X  
DOI: <https://doi.org/10.1016/j.apsusc.2018.06.226>  
Reference: APSUSC 39734

To appear in: *Applied Surface Science*

Received Date: 1 March 2018  
Revised Date: 11 June 2018  
Accepted Date: 25 June 2018

Please cite this article as: L. Scholtz, P. Šutta, P. Calta, P. Novák, M. Solanská, J. Müllerová, Investigation of barium titanate thin films as simple antireflection coatings for solar cells, *Applied Surface Science* (2018), doi: <https://doi.org/10.1016/j.apsusc.2018.06.226>

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# Investigation of barium titanate thin films as simple antireflection coatings for solar cells

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## Abstract

Barium titanate (BaTiO<sub>3</sub>, BTO) is a perovskite class material of remarkable dielectric, ferroelectric and ferromagnetic properties. Our previous studies on optical properties of BTO thin films proved high visible transmittance and sharp absorption edge at ~ 300 nm. Therefore the usage of BTO as a UV blocker or an antireflection (AR) coating in visible region is straightforward. AR coatings are agreed to be important parts of many photonic devices, among them also of solar cells. In this paper, single layers of amorphous BTO are numerically and experimentally investigated as promising AR coatings for achieving increased light trapping in thin film silicon solar cells. Reduced reflections achieved by BTO thin films deposited using RF magnetron sputtering on a-Si:H/SiO<sub>2</sub> compared with pristine a-Si:H/SiO<sub>2</sub> system are clearly demonstrated. Antireflection effects are analyzed using simple AR systems comprising BTO.

*Keywords: antireflection, AR coatings, barium titanate, thin films, reflectance efficiency*

## 1. Introduction

Low-cost, simple and industry friendly procedures and materials for solar cells are still the main questions for many researchers groups. Since silicon solar cells technologies are already close to their physical limitations there is still necessity of new structures or materials as combination of various types of solar cell technologies, i.e. promising perovskite solar cells [1]. One way to increase the effectivity is to reduce optical loss and to achieve effective light trapping, e.g. via antireflection (AR) coatings and/or the micro-scale texturing of cell surfaces [2, 3, 4, 5]. Due to promising optical properties perovskites bring hope not only as solar absorbers, but some of them also as supporting optical layers, i.e. for antireflection. Perovskite titanium oxides have already been reported to operate as effective AR single layer coatings for silicon solar cells [6].

Titanium-based mixed-metal perovskite oxides of the chemical formula A<sup>II</sup>Ti<sup>IV</sup>O<sub>3</sub> with A<sup>II</sup> being great metal cations manifest unique physical and chemical properties [7, 8]. Chemical compounds with Ca, Pb, Sr, Ba, Zn, Fe etc. used as A element have outstanding potential in dielectric and opto-electronic applications, e.g. transparent electronics, fuel cells, gas sensors, memory devices, photocatalysis, non-linear optics, solar energy conversion for water splitting and decontamination [9, 10, 11]. The same octahedral coordination evokes the similarity of optical properties. Optical properties of ZnTiO<sub>3</sub>, SrTiO<sub>3</sub> (STO) and BaTiO<sub>3</sub> (BTO) were studied in details [12, 13, 14]. In general titanium-based oxides are ferroelectrics of wide-band gaps but the band gaps may differ substantially in dependence on structure, microstructure and composition. For this reason, although having been studied recently as

thin films or nano-particles [15, 16, 17, 18] optical properties of BTO have been far from complete for the time being. Special attention has been given to absorptive properties and band gaps [19, 20, 21]. Motivations for progressive optical and electro-optical studies are e.g. the applications as nano-photonic electro-optic devices [22, 23].

BTO thin films are distinguished by broad UV Vis transparency and relatively high refractive index  $> 2.0$  (589 nm) [6, 24, 25]. Therefore similarly to well-known high-refractive index oxides, e.g. ZnO, ZrO<sub>2</sub>, TiO<sub>2</sub>, Ta<sub>2</sub>O<sub>5</sub>, the application in AR coatings can be anticipated. Spectral regions of their transparency compare well with the solar irradiance spectrum. Therefore concepts of trapping light in solar cell absorbers using BTO AR coatings could be promising. Improved light-trapping in solar cells using a single layer [6] or multilayer ferroelectrics have already been reported [26, 27]. Benefits of using ferroelectric materials as AR coatings come from their relatively simple technological preparation and the sensitivity of their physical properties to external effects, e.g. ambient conditions or external electric field.

In this paper we examined the usage of single and simple multilayer BTO thin films as AR coatings for solar cells. AR effects of simple BTO structures on a-Si:H/SiO<sub>2</sub> and SiO<sub>2</sub> substrates were studied. As it is well-known that optical properties of thin films are deposition process and deposition conditions dependent, prior to the AR analysis optical properties of BTO thin films (refractive indices and extinction coefficients) deposited by RF magnetron sputtering on SiO<sub>2</sub> substrate were extracted from transmittance spectra. Subsequently these optical properties were used to numerically design specific AR coatings. Simulation results were used for exact AR coatings depositions and experimental verifications.

## 2. Experimental section

BTO thin films were deposited from BaTiO<sub>3</sub> ceramic target (purity 99.99%) by 13.56 MHz RF magnetron sputtering in argon atmosphere using BOC Edwards TF 600 coating system. More details can be found in [28]. The substrate temperature was kept at 350 °C. The samples were not post-deposition treated. Depositions of two BTO films of the intended thickness of 50 and 100 nm were completed on two substrates:

- BTO deposited on SiO<sub>2</sub> labelled as No.1 (intended thickness of ~ 50 nm) and No.2 (intended thickness of ~100 nm) used for the determination of the optical properties and for considering AR design,
- BTO (No.3 and No.4) of the approximate thicknesses as above deposited on thin amorphous hydrogenated silicon (a-Si:H) on SiO<sub>2</sub> used for structure analysis and for considering AR design.

Thin film structure has an evident impact on BTO optical properties a therefore should be known in all light-based applications of BTO. As shown in [29] after annealing crystalline titanates manifest increased extinction coefficients as well as refractive indices. Therefore the X-Ray diffraction analysis of as-deposited samples on SiO<sub>2</sub> and crystalline Si (c-Si) was performed. BTO XRD pattern were taken by an automatic powder diffractometer XPert Pro equipped with a proportional point detector and CuK<sub>α</sub> radiation as an X-ray source. Seemann-Bohlin asymmetric goniometer arrangement with the fixed angle of X-rays incidence was used [28]. In Fig.1 XRD patterns are introduced to confirm amorphous nature of BTO thin films presented by broad bands in 2  $\theta$  scales no matter which substrate was used. XRD patterns of BTO of intended thickness of 50 nm and 100 nm deposited on SiO<sub>2</sub> and c-Si

manifest broad angle asymmetry caused by the composition of amorphous SiO<sub>2</sub> and BTO diffraction. Sharp lines are Laue diffraction lines resulting from c-Si substrate.

Optical transmittances at nearly normal incidence of the samples No.1 and No.2 were recorded in wide spectral range of (250 – 1100) nm by Specord 210 spectrophotometer with air blank reference channel. Transmittance measurements were used for the analysis of optical properties of BTO. Additional transmittance/reflectance spectra were recorded for the sake of considering AR effects. Oblique reflectance measurements were performed at the 45 angle of incidence.

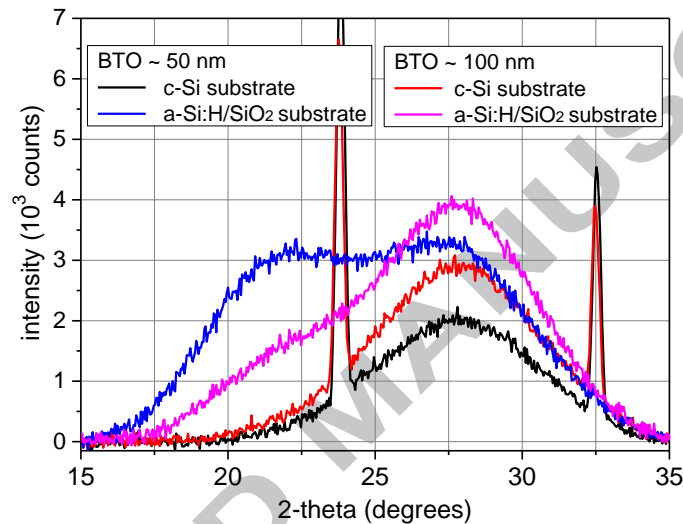


Fig.1. XRD patterns of the as-deposited BTO thin films of the thickness of ~ 50 nm and ~ 100 nm on indicated substrates. The vertical scale is reduced to highlight BTO patterns and suppress sharp high Laue peaks from c-Si substrate.

### 3. Optical properties of BTO thin films for prospective AR coatings

Although a single AR coating usually does not work in spectral broadband, any improvement in reflectance reduction of a solar cell upper surface is welcome when using the simplest technological practice, i.e. a single AR coating.

The main analytical design rule for a single-layer AR coating is destructive interference in the reflected light which can be achieved at the wavelength  $\lambda$  using the quarter-wavelength thickness (QWOT) of the layer  $d_{QWOT} = \frac{\lambda}{4n}$  where  $n = \sqrt{n_1 n_2}$  and  $n_1$ ,  $n_2$  are refractive indices of the layer and the ambient, usually glass cover in solar cells ( $n_2 \sim 1.4$ ). Let us use the design wavelength  $\lambda = 550$  nm. At this wavelength BTO does not absorb light. For brief initial design of single AR coatings the BTO refractive index was taken from [13] ( $n_1 \sim 2$ ). Then the single AR BTO thickness should be ~ 80 nm.

AR design requires knowledge of the exact complex wavelength-dependent refractive index  $N(\lambda) = n(\lambda) + ik(\lambda)$  where the real part  $n$  is known as the refractive index and  $k$  as the extinction coefficient expressing optical loss in optical medium. Information on the absorptive properties is important due to the fact that an AR coating should absorb the incoming light as little as possible. It is well-known that the optical properties of thin films may depend on the deposition chemistry, deposition conditions and even the thin film thickness. Therefore prior

to AR numerical design the refractive indices and extinction coefficients of BTO thin films must be determined as depend on the film thickness. Following the above recommendation on the AR coating thickness two BTO thin films No.1 and No.2 of the intended thickness 50 nm and 100 nm were deposited.

Refractive indices and extinction coefficients are usually deduced from reflectance/transmittance spectra. Transmittance of a homogeneous thin film with parallel interfaces deposited on a thick substrate is a nonlinear function of the wavelength, refractive indices and extinction coefficients of the film and substrate and of the film thickness. Refractive indices and extinction coefficients of the films were extracted from the measured transmittance using an optimization fitting procedure based on a genetic algorithm. The theoretical transmittance to be compared with the experiment was calculated using the theory in [30] and the Tauc-Lorentz dispersion model currently employed for the parameterization of the optical functions of amorphous semiconductors [31].

Experimental transmittances of the samples of No.1 and No.2 can be seen in Fig. 2 (solid lines). Few interference fringes are seen in spectra due to weak absorption of light above the absorption edge. The theoretical fitted transmittance calculated using the Tauc-Lorentz model is included (dash lines). The refractive indices and extinction coefficients are in Fig. 3.

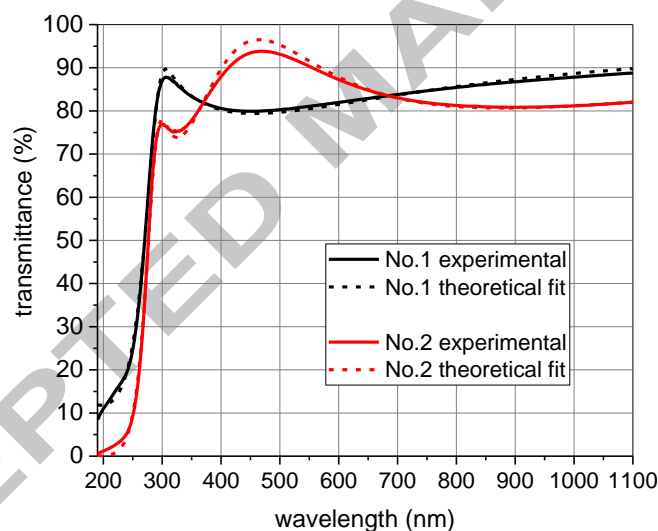


Fig.2. Experimental transmittance of BTO thin film No.1 and No.2 on SiO<sub>2</sub> substrate. Theoretical fitted transmittance is included.

The transparency onset is steep at  $\sim 300$  nm although is partially shifted to higher wavelengths for the thicker BTO film. Negligible extinction coefficients for  $\lambda > 300$  nm (Fig. 3) confirm the desirable BTO transparency in UV Vis region. Therefore this material does not involve additional absorption loss in solar cells when used as an AR coating.

The differences in optical properties of BTO of varied thickness correspond well to the discoveries reported in [24]. The refractive index increases with increasing thickness, although the differences between the thinner No.1 and thicker No.2 samples are negligible in the non-absorbing region. The thickness of the films as one of the fitting parameters of the optimization fitting procedure was established to be 61 nm (sample No.1) and 116 nm (sample No. 2).

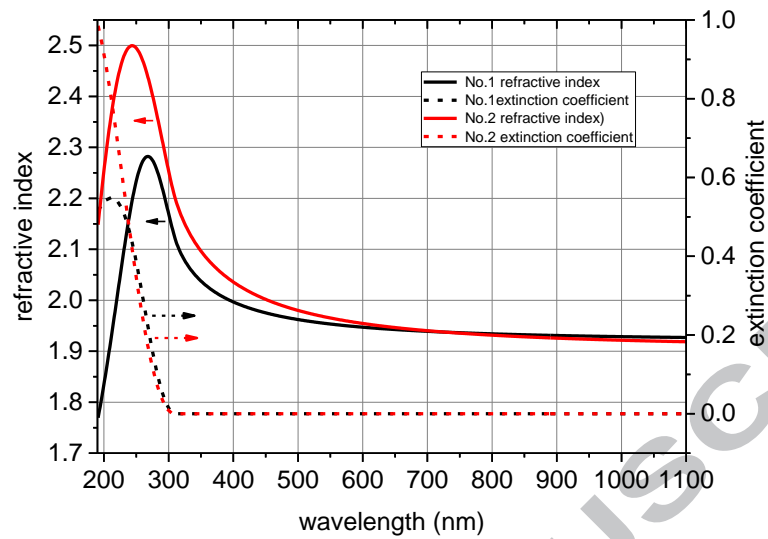


Fig.3. Refractive indices and extinction coefficients of the samples No.1 (~ 50 nm) and No.2 (~ 100 nm).

#### 4. Results and discussion

Using collected knowledge on refractive indices and extinction coefficients of BTO thin films we numerically calculated several structures comprising BTO and consider their AR properties. Subsequently we compare numerical results with experimental AR performance of BTO of similar structure as simulated.

##### 4.1. Simulations of single-layer AR coatings

To get a broader vision the simulations of BTO AR coatings as single or multiple layer structures were performed using the transfer matrix method [32,33].

As mentioned above single-layer AR coatings usually do not work in spectral broadband. Adopting optical properties of BTO obtained from our experimental analysis, reflectance and transmittance spectra of various single layer BTO AR coatings at nearly normal incidence were calculated. The optical properties of  $\text{SiO}_2$  and a-Si:H used as the substrates usually present in amorphous silicon thin film solar cells were also taken from our experimental analysis [30]. In Fig. 4 and Fig. 5 simulated reflectances and transmittances at nearly normal incidence of BTO deposited on a-Si:H/ $\text{SiO}_2$  can be seen. In simulations 230 nm a-Si:H thin film on thick incoherent  $\text{SiO}_2$  substrate was considered.

Although realizing the dependence of optical parameters of thin films of the thickness, for simplification BTO films of the thickness 80 nm were supposed to have optical properties as No.1 sample. BTO films of higher thickness were simulated with the optical properties of No.2 BTO sample. This simplification is granted by the basic message of this investigation – showing the possibility to use BTO for AR purposes.

For the comparison the reflectance and transmittance of the pristine a-Si:H/ $\text{SiO}_2$  structure is indicated. As a-Si:H/ $\text{SiO}_2$  is specified as the substrate for BTO, a-Si:H of the thickness of 230 nm typical for a-Si:H solar cells is actually a thin film on  $\text{SiO}_2$ . Therefore interference features are apparent in the reflectance and transmittance spectra of a-Si:H/ $\text{SiO}_2$ .

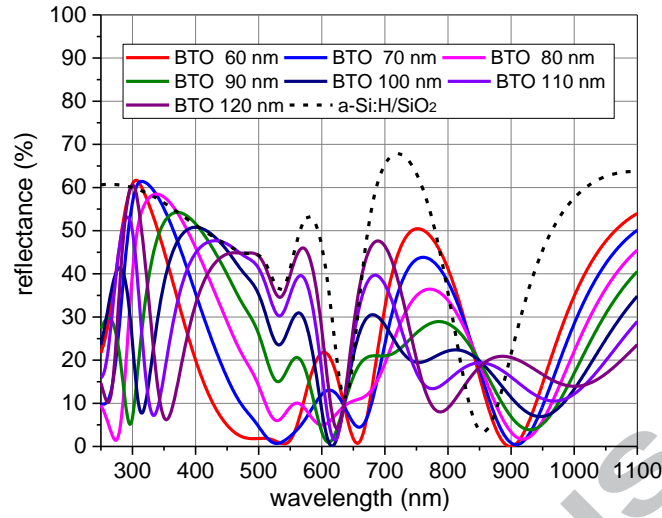


Fig. 4. Nearly normal reflectances of single layer BTO AR coatings on a-Si:H/SiO<sub>2</sub> substrates. The varied thickness of BTO thin films is indicated.

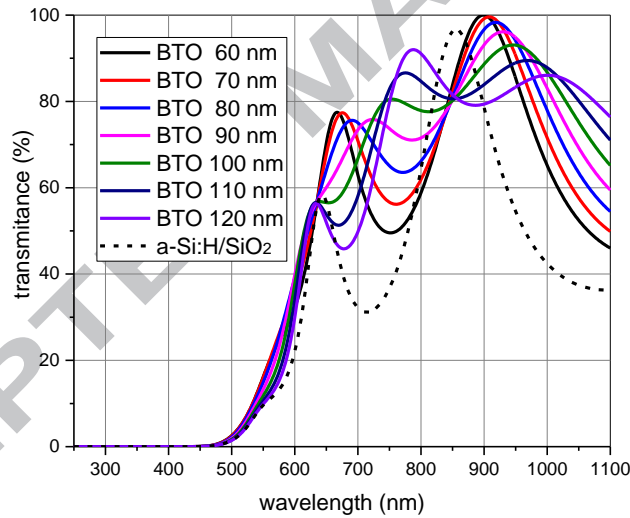


Fig. 5. Nearly normal transmittances of single layer BTO AR coatings on a-Si:H/SiO<sub>2</sub> substrates. The varied thickness of BTO thin films is indicated.

It is obvious that in case of a single AR coating the reflectance reduction is not broadband and often not straightforward. But still we hope to achieve an integral reflectance reduction within a certain spectral range. To identify AR performance we define the AR reflectance efficiency  $\eta_R$  using the relative integral reflectance decrease within a specific spectral range (similarly to [34])

$$\eta_R = \frac{\int R_0(\lambda)d\lambda - \int R(\lambda)d\lambda}{\int R_0(\lambda)d\lambda} \cdot 100\% \quad (1)$$

where  $R(\lambda)$ ,  $R_0(\lambda)$ , are the reflectances with and without an AR coating, respectively. Similarly we can define the transmission efficiency that is expected also to attain enhanced values using an AR coating

$$\eta_T = \frac{\int T(\lambda)d\lambda - \int T_0(\lambda)d\lambda}{\int T_0(\lambda)d\lambda} \cdot 100\% \quad (2)$$

where  $T(\lambda)$ ,  $T_0(\lambda)$ , are the transmittances with and without an AR coating, respectively. Apparently for effective antireflections  $\eta_R$  and  $\eta_T$  should increase.

Solar cells often do not optimally face the sun and usually experience many hours of oblique sunlight incidence. The sensitivity to oblique angles of incidence is an important issue. Therefore besides nearly normal transmittances and reflectances of single BTO layer on a-Si:H/SiO<sub>2</sub> also transmittances and reflectances at 45° angle of incidence were simulated as polarization average.

Integral reflectances and transmittances used in Eqs. (1) and (2) were calculated over the spectral range from 250 to 1100 nm. In Fig.6 reflectance and transmittance efficiencies at nearly normal and 45° angle of incidence are depicted for BTO of different thickness on a-Si:H/SiO<sub>2</sub>.

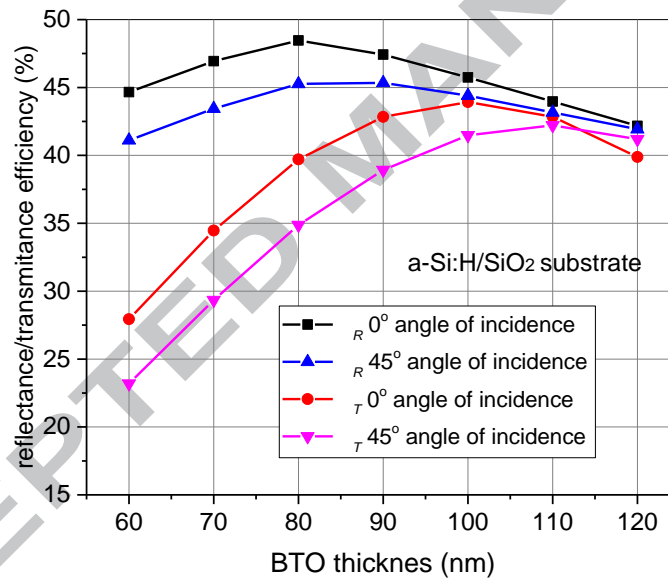


Fig. 6. Reflectance and transmittance efficiencies of single layer BTO AR coatings on a-Si:H/SiO<sub>2</sub> substrates as a function of varied thickness of BTO thin films.

As predicted we conclude that the best AR efficiency  $\eta_R$  at both angles of incidence can be achieved for ~ 80 nm BTO. In comparison with the pristine a-Si:H/SiO<sub>2</sub> nearly normal reflectance reduction of ~ 45 % can be achieved. The transmittance efficiency reaches the maximum for ~ 100 - 110 nm BTO layer. The shift is caused by the absorption in a-Si:H. Of course following the BTO antireflective mission, the reflectance efficiency is more important for AR applications. The differences in  $\eta_R$  within the simulated thickness range are not significant which is a good message for facilitating the thickness control at BTO deposition.

#### 4.2. Simulations of multiple-layer AR coatings

If more layers than one are used in AR coatings their composition and thickness must be optimized to give a broader AR band. Therefore our further scenario was to simulate



reflectance of simple double AR coatings deposited on a-Si:H/SiO<sub>2</sub>. Reflectance efficiencies for nearly normal incidence are in Fig. 7 for the structures xSiO<sub>2</sub>/xBTO and xBTO/xSiO<sub>2</sub> where  $x$  is the layer thickness. BTO was simulated as the high-index material, SiO<sub>2</sub> as the low-index material. We see that the xSiO<sub>2</sub>/xBTO structure offers higher reflectance efficiencies. No less noticeable observation is that the reflectance efficiencies of xSiO<sub>2</sub>/xBTO are over 50 % for a wide range of thicknesses and the maximum of reflectance efficiency is very mild. Therefore fewer constraints on the exact layer thickness at the potential deposition are laid.

We see that double layer AR coatings offer similar reflectance efficiencies than single BTO layers but can be achieved with thinner layers of BTO. We conclude that one BTO layer or SiO<sub>2</sub>/BTO structure are enough to decrease reflection loss by half. However it must be emphasized that highly sensitive optimization concerning the AR structure composition and layer thickness is remarkably meaningful. We see that antireflection of the double structure is more sensitive to oblique incidence of 45° than a single layer.

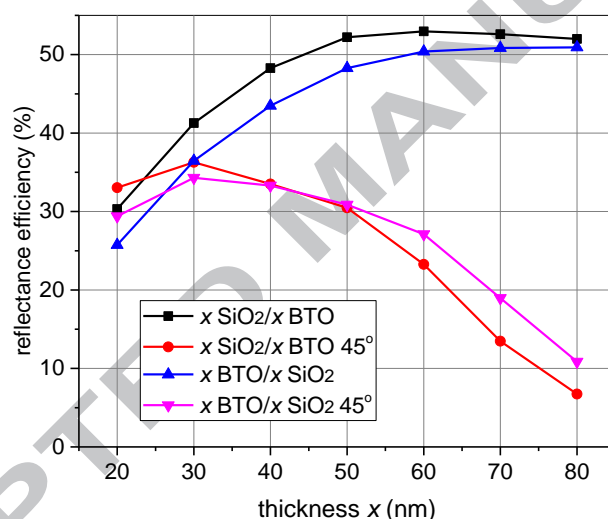


Fig.7. Nearly normal and oblique (45°) reflectance efficiencies of double AR coatings comprising BTO and SiO<sub>2</sub> of the thickness  $x$  on a-Si:H/SiO<sub>2</sub>/substrates.

In the next course of simulations AR coatings consisting of additional layers were examined. Although being more complex related to single BTO AR coatings these structures remain still very simple in comparison with typical broadband AR coatings comprising tens of layers. Multi-layered structures with periodical SiO<sub>2</sub>/BTO of varied thickness in the middle of the structure sandwiched between the ambient and SiO<sub>2</sub> substrate were simulated. Pure SiO<sub>2</sub> substrate was selected here because in the case of the superstrate a-Si:H solar cells the upper layer of the cell is glass or SiO<sub>2</sub> with transparent conducting oxide layer (so-called TCO) below followed by a-Si:H solar cell absorber. The reflectance of 50 nm thin film BTO on SiO<sub>2</sub> was used as the reference to be compared with more complex structures.

Fig.8 depicts the selection of the best representatives. The most prospective results were obtained with 30 nm BTO/(30 nm SiO<sub>2</sub>/30 nm BTO) <sup>$m$</sup>  and 20 nm BTO/(20 nm SiO<sub>2</sub>/20 nm BTO) <sup>$m$</sup>  (the repetition number  $m = 3$  or 5). The reflectance efficiencies are given in Fig.8. Nevertheless the reflectance efficiency of 48.6 % by 20 nm BTO/(20 nm SiO<sub>2</sub>/20 nm BTO)<sup>5</sup> is

still under the best  $\eta_R$  achieved for single BTO or double  $\text{SiO}_2/\text{BTO}$  layers. With respect to these results we conclude that additional layers do not bring any notable benefit and single or double layers are more antireflective efficient.

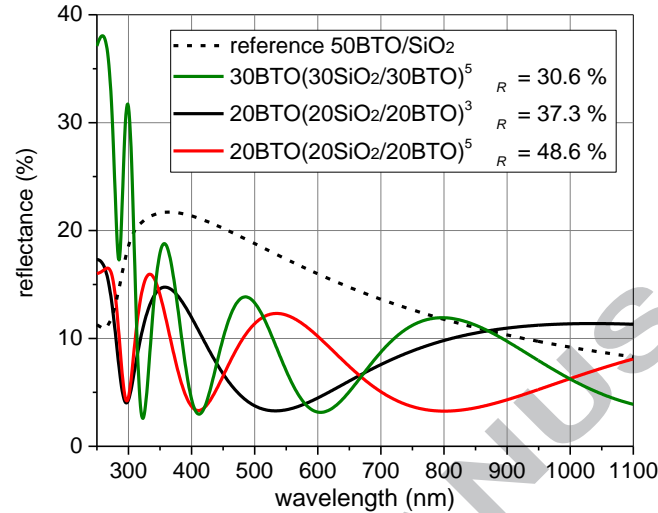


Fig. 8. The comparison of the nearly normal reflectance of multilayer BTO structures with single 50 nm BTO layer on  $\text{SiO}_2$ . The numbers in the legend indicate the layer thickness (nm).

#### 4.3. Comparison of simulation results with some experimental AR coatings

Partial experimental verification to support the idea of suitability of BTO in AR effects is based on No.1 and No.2 samples deposited on  $\text{SiO}_2$  (see Section 3.1 and relating Figs. 2 and 3) and No. 3 and No. 4 deposited on  $\text{a-Si:H/SiO}_2$  and intended to differ by the films thickness similarly as No.1 and No.2., i.e.  $\sim 60$  and  $\sim 110$  nm (Table 1). The thickness of  $\text{a-Si:H}$  deposited on  $\text{SiO}_2$  to form  $\text{a-Si:H/SiO}_2$  substrate for BTO (No. 3 and No. 4) was determined by the numerical method described in the Section 3.1 to be of 230 nm.

Transmittances and oblique reflectances at 45 angles of incidence were recorded (Figs. 9 and 10). In all plots corresponding transmittances/reflectances of the substrates are depicted. The reflectance/transmittance efficiencies were calculated as related to the integrated reflectance/transmittance of the specific  $\text{SiO}_2$  and  $\text{a-Si:H/SiO}_2$  substrates. Experimental transmittance/reflectance efficiencies and those calculated from simulated spectra are in Table 1. Both single BTO AR coatings on pure  $\text{SiO}_2$  (No.1 and No.2) are useless not enabling to achieve antireflections. However their experimental and simulated transmittance efficiencies  $\eta_{Texp}$  and  $\eta_{Tsimul}$  are in acceptable agreement. The differences in transmittance/reflectance efficiencies of BTO on  $\text{a-Si:H/SiO}_2$  are probably due to the overestimation of the experimental film thicknesses of  $\sim 60$  nm and  $\sim 110$  nm.

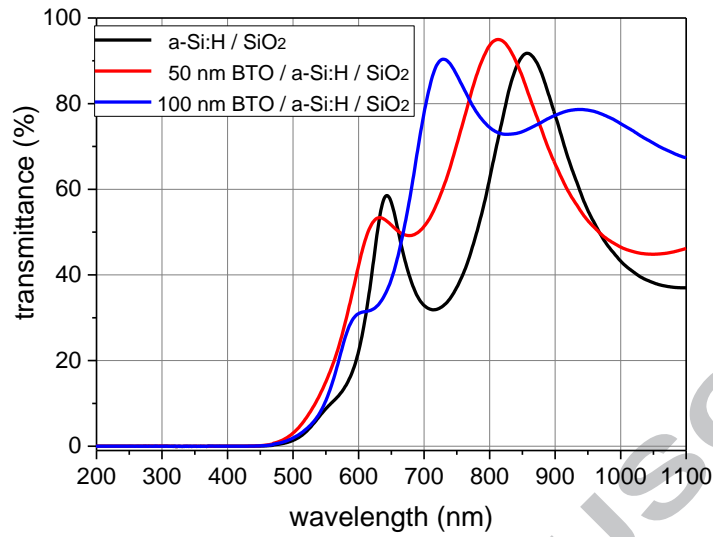


Fig. 9. Normal experimental transmittance of BTO thin films on a-Si:H/SiO<sub>2</sub> substrate. Transmittance of the substrate indicated for the comparison (black line).

Table 1. Experimental single BTO AR coatings of the specified thickness  $d$  deposited on indicated substrates. Transmittance efficiency of samples on SiO<sub>2</sub> calculated from Fig. 2.

No.	Substrate	$d$ (nm)	$\eta_{Texp}$ (%)	$\eta_{Tsimul}$ (%)	$\eta_{Rexp}$ (45° of incidence) (%)	$\eta_{Rsimul}$ (45° of incidence) (%)
1.	SiO <sub>2</sub>	61	15.8	14.12		
2.	SiO <sub>2</sub>	112	17.5	15.09		
3.	a-Si:H/SiO <sub>2</sub>	~ 60	18.9	27.94	35.3	41.11
4.	a-Si:H/SiO <sub>2</sub>	~ 110	34.4	42.85	38.8	43.16

## Conclusions

In this paper we numerically and experimentally investigated the usage of amorphous BTO thin films to work as simple AR coatings. This usage may help in reducing the reflection loss and in improving the light trapping in thin film silicon solar cells. Reduced reflections represented by the reflectance efficiency achieved by BTO thin films on a-Si:H/SiO<sub>2</sub> compared with pristine a-Si:H/SiO<sub>2</sub> system are clearly demonstrated. We conclude that one BTO layer or SiO<sub>2</sub>/BTO structure are enough to decrease reflection loss by half. Numerical experiments with multilayer AR systems comprising BTO and SiO<sub>2</sub> does not show improvements. Therefore no specific multilayer AR coatings demanding sophisticated design and technology are necessary to achieve success. Our results demonstrate that simple AR coatings of BTO can be functional in reducing reflectance loss in solar cells. However a thorough optimization of the film thicknesses in the single and double AR design is recommended.

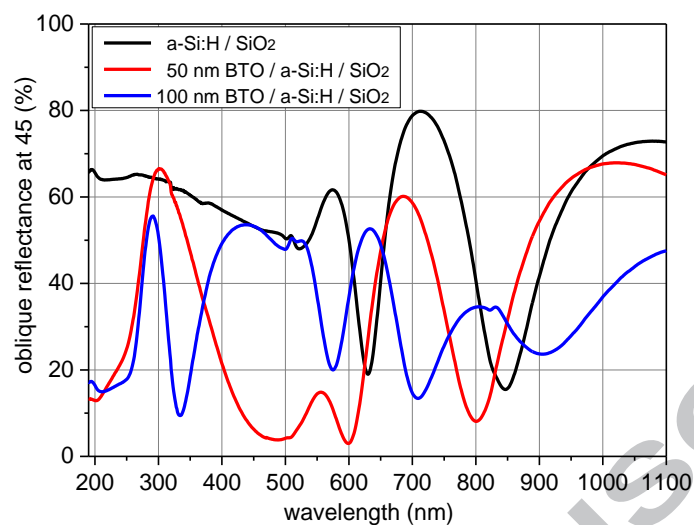


Fig. 10. Oblique experimental reflectance of BTO thin films on a-Si:H/SiO<sub>2</sub> substrate (45° angle of incidence). Reflectance of the substrate indicated for the comparison (black line).

### Acknowledgements

This work was partly supported by the Slovak Research and Development Agency under the projects APVV-0888-11 and APVV-15-152. The results were partially developed within the CENTEM Project, reg. no. CZ.1.05/2.1.00/03.0088 co-funded by the ERDF within the OP RDI Programme, and in the follow-up sustainability stage, supported through CENTEM+ (LO1402) by financial means from the Ministry of Education, Youth and Sports of the Czech Republic under the National Sustainability Programme I.

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**Highlights**

RF magnetron sputtered thin films of barium titanate

Exclusively sharp absorption edge

Barium titanate as a single antireflective coating on silicon and glass

Antireflection efficiency analyzed theoretically and experimentally

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