



**EQE Analysis/Simulation Program**

*e-ARC Software*

*Version 1.1*



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## About Version 1.1

The e-ARC software Version 1.0 was released in September 25, 2017 at EU PVSC conference. Later, however, it was found that the software does not operate properly in European-language-based Window OS due to the difference in the definition of “comma” and “period”. In this new version of the software (Version 1.1), the problem has been fixed and the operation of the software under various Window operating systems has been confirmed. Moreover, simulation/analysis examples for different solar cell devices are included (see “2.3 Quick start”).

If there are some problems for the software operation, please contact with the following persons:

i) Concerning overall software operation

Hiroyuki Fujiwara (Gifu University); Email: fujiwara@gifu-u.ac.jp

ii) Concerning software download

Hitoshi Tampo (AIST) ; Email: tampo-21@aist.go.jp

## 1. Introduction

The e-ARC software is the external quantum efficiency (EQE) analysis and simulation program that can be employed to determine reflection, absorption and recombination losses in various thin film solar cells with a calculation time of ~1 s. In this method, the effects of (i) light scattering caused by rough textures in submicron ranges and (ii) carrier loss induced by recombination at semiconductor interface and bulk regions are fully incorporated. In the ARC method, the reflectance ( $R$ ) spectrum of the textured solar cell is calculated by linearly connecting the minimum  $R$  points obtained assuming flat optical models to simulate the anti-reflection condition (ARC) effect realized in textured structures. In the extended ARC (e-ARC) method, the influence of the carrier recombination is further modeled. The most detailed explanation for the analysis method is described in the book

- [1] Chapter 2 in *Spectroscopic Ellipsometry for Photovoltaics Volume 2: Applications and Optical Data of Solar Cell Materials*, H. Fujiwara and R. W. Collins, editors, Springer: to be published in 2018.

In this book, the tabulated and modeled optical constants of ~150 solar-cell component

materials including inorganic/organic semiconductors, transparent conductive oxide (TCO) materials, metal, and substrates are summarized, which can be incorporated directly to the EQE calculations. The basic principles of the EQE analysis method can also be found in

- [2] A. Nakane, H. Tampo, M. Tamakoshi, S. Fujimoto, K. Min Kim, S. Kim, H. Shibata, S. Niki and H. Fujiwara, J. Appl. Phys. **120** (2016) 064505.

In the above Ref. [2], the EQE analysis examples of  $\text{Cu}_2\text{ZnSn}(\text{S},\text{Se})_4$  (CZTSSe), CdTe, hybrid perovskite solar cells are described. For the complete analysis of  $\text{Cu}(\text{In},\text{Ga})\text{Se}_2$  (CIGSe) solar cells fabricated by three-stage coevaporation, the method described in

- [3] T. Hara, T. Maekawa, S. Minoura, Y. Sago, S. Niki and H. Fujiwara, Phys. Rev. Applied **2** (2014) 034012.

has been employed in the software.

When the analysis/simulation results obtained using the e-ARC software is to be published in your researches, we would like to ask you to cite the proper references described above.

## 1.1 Software features

The e-ARC software calculates the EQE and absorptance ( $A$ ) spectra of the semiconducting absorber and component layers incorporated into thin-film solar cells in high speed. From these spectra, the short-circuit current density ( $J_{\text{sc}}$ ) and the current losses induced by light reflection, parasitic light absorption and carrier recombination are estimated.

## 1.2 Software specification

### ① Modeling and calculation

- Thin-film solar cells with substrate- and superstrate-type structures can be calculated.
- CIGSe solar cells having Ga double-grading structures can be simulated.
- By linear interpolation, various EQE and  $R$  spectra with different data spacing for wavelength can be analyzed.
- Arbitrary optical data (tabulated and modeled data) can be used as the optical constants of each layer. The model parameters of the Tauc-Lorentz, Drude, Sellemier and Cauchy models can also be used. For the details of the models, please see Ref. [1] or [4] H. Fujiwara, *Spectroscopic Ellipsometry: Principles and Applications*, Wiley, 2007.

- Minor adjustment of band gap ( $E_g$ ) of each layer can be made quite easily by using a simple spectral-shift model.
- Unfortunately, in the current version, the EQE analyses of wafer-based (thick) solar cells are not supported. This analysis feature will be provided in ver. 2.0 which will be released in 2018.

## ② Analysis results

- $R$ ,  $A$  and EQE spectra of solar cells are calculated in a few seconds by applying the optical admittance method assuming flat optical models.
- $J_{sc}$  values corresponding to EQE,  $R$ ,  $A$  and recombination are shown automatically.
- When the thickness of the component layer is changed, all the spectra changes automatically (real-time mode).
- Partial EQE and  $A$  spectra (depth-resolved EQE and  $A$  spectra) can be calculated.

## ③ File management

- All the graphs that appear on the software screen can be saved using “.png” file format.
- All the calculation parameters and results can be saved.
- The partial EQE and  $A$  data (2D data) can be saved using a script file that can be illustrated by gunplot.

## 2. Installation & Quick Start

### 2.1 Execution environment

- Microsoft Windows 7 and above operating system
- For gunplot (free software), gnuplot 4.2 and above version

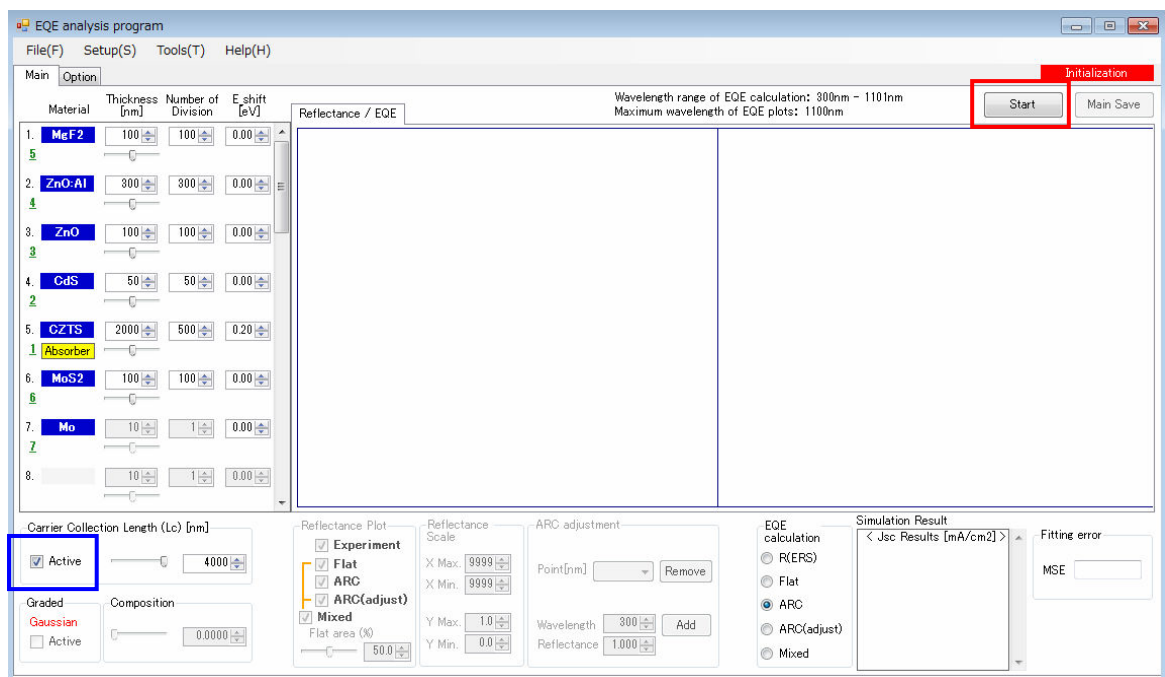
### 2.2 Installation method

The easiest way to operate the software quickly is to copy the downloaded “e-ARC” folder directly to the main C drive (i.e., root directly; C:). To do this, click “e-ARC.zip” and copy the “e-ARC” folder to the C drive (C:). Note that when the e-ARC folder is copied to the desktop and “C:/Program” directory, the software does not operate properly. In case the e-ARC folder is copied to other directory, the file path name should be changed (see Sections 4.1 and 4.2). After the completion, click “Optical simulator

ver1.1.exe” in the e-ARC folder to start the program (If the e-ARC folder is copied to the directory other than C:, some error messages appear due to the improper path names, which need to be corrected).

## 2.3 Quick start

The window below shows the screen image obtained after clicking “Optical simulator ver1.1.exe”. The initial setup is for CZTS solar cell. If you click the start button indicated by a red square below, the EQE spectrum is calculated.

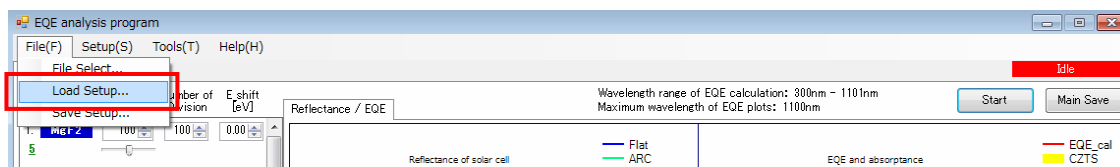


If the layer thickness is changed, the EQE spectrum changes automatically. However, please deactivate the carrier collection length correction (blue square above) if the recombination analysis is not necessary (Default setup is “Active”).

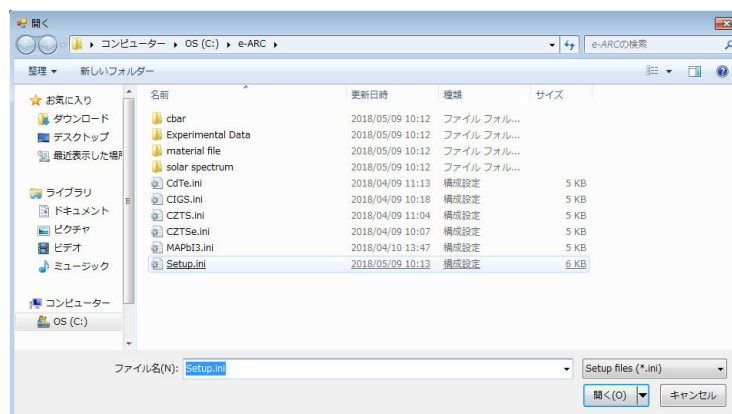
In the software, several examples for different device structures are installed. To load this setup, first click the stop button shown below.



Then click the “File” menu bar and select “Load Setup”.



When the following sub-window opens, select a file with “.ini” extension.



The files with “.ini” extension indicate calculation examples and, in the current version, the examples for CdTe (i.e., “CdTe.ini” for example), CIGS, CZTS, CZTSe, and MAPbI<sub>3</sub> solar cells are included. After selecting these files, click the start button to start the calculation. The current setup can also be saved using “Save Setup” command in the “File” menu. Note that “.ini” files need to be saved in the main e-ARC directory.

## 2.4 Directory structure

### 📁 e-ARC

- 📁 cbar: Color examples for 2D graphs (default)
- 📁 material file: Optical constants of materials are stored
- 📁 Experimental data: Experimental EQE and *R* spectra of analysis examples
- 📁 solar spectrum: Spectrum of solar light (AM1.5G) is stored
- Optical Simulator ver1.1.exe: Execution file of the e-ARC software (Version 1.1)
- Setup.ini: Initial parameter file for the program
- ..... ini: Several initial files for different solar cells (see above)

### 3. Overview of EQE analysis/simulation

Figure 3.1 summarizes the EQE analysis procedure in the software. In the analysis, we first select experimental data of the solar light spectrum, EQE spectrum (optional) and  $R$  spectrum (optional). The experimental EQE spectrum is necessary when the fitting analysis is made and is unnecessary for simulations, whereas the experimental  $R$  spectrum can be incorporated into the analysis particularly when the texture structure is present and the modeling of accurate  $R$  spectra is difficult (see Ref. [1,2]). In the next step of the analysis, the layer structure of the solar cell is modeled, followed by the selection of the optical constants and thickness of each layer. After setting these initial parameters, the calculation is started. In the actual fitting analysis, layer thicknesses and carrier recombination (carrier collection length  $L_C$  and dead layers: see Section 4.6 and 4.7) can be adjusted. In more advanced analysis, the compositional grading in the absorber layer is modeled.

In the software, mean-squared error (MSE) given by the following equation is calculated automatically:

$$MSE = \frac{1}{N} \sum_{i=1}^N (EQE_{ex,i} - EQE_{cal,i})^2, \quad (3.1)$$

where  $EQE_{ex}$ ,  $EQE_{cal}$  and  $N$  are experimental EQE and calculated EQE and a total number of data points, respectively. When the fitting is insufficient (i.e., when a MSE value is large), the structures and optical constants of layers are adjusted. Finally, from the best fitting, the current losses are determined.

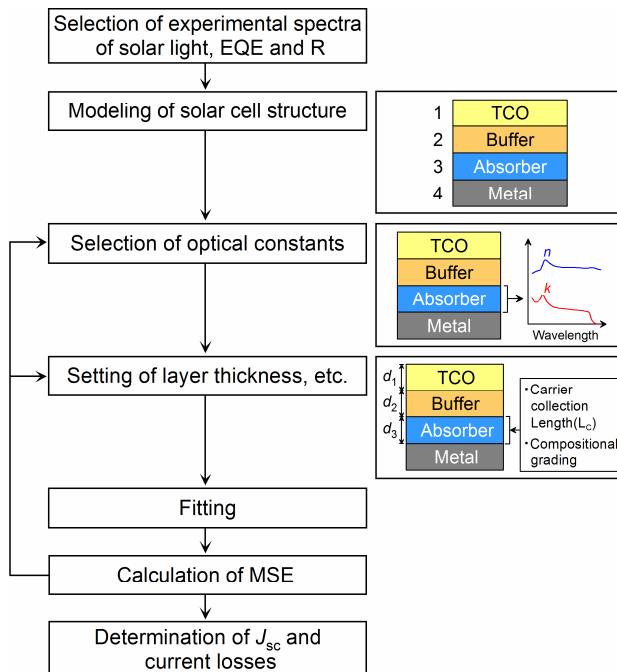


Fig. 3.1 EQE calculation procedure.



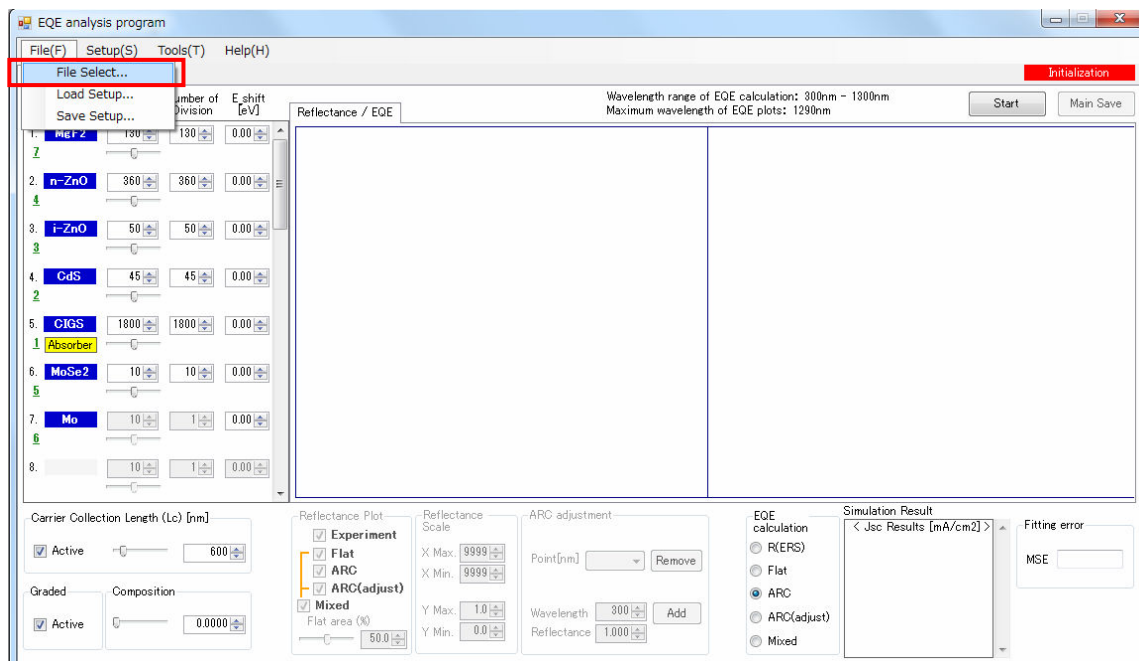
#### 4. Example 1: EQE analysis of a substrate-type CZTSe solar cell

In this section, the basic e-ARC analysis is described using a CZTSe solar cell as an example. The experimental data are described in Ref. [2]. This CZTSe solar cell is analyzed assuming a structure of ZnO:Al/ZnO/CdS/CZTSe/MoSe<sub>2</sub>/Mo (a total of six layers).

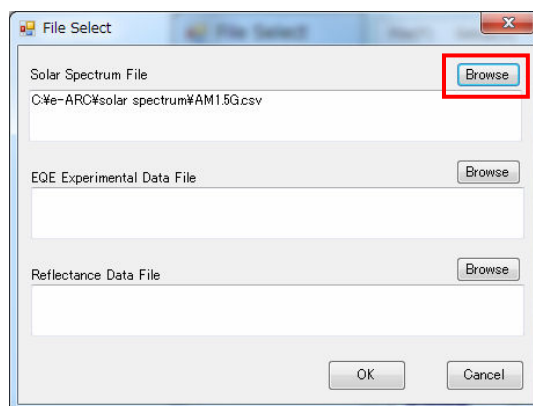
##### 4.1 Selection of the experimental data and path name setting

In this analysis example, the  $R$  spectrum of a CZTSe solar cell is calculated by the ARC method [3] as the solar cell has a submicron texture [2]. In the analysis, therefore, only the solar light and EQE spectra are used. The unit of the input solar cell spectrum is  $\text{Wcm}^{-2}\text{nm}^{-1}$  and the spectra of EQE and  $R$  should be in a range of 0~1 (i.e., Not 0-100%).

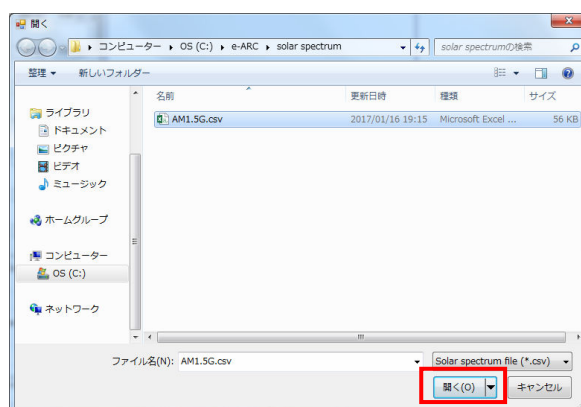
- ① Start the software by clicking “Optical Simulator ver1.1.exe”.
- ② The software window below shows the initial screen of the software. Click the tab menu of “File” and further select “File Select”



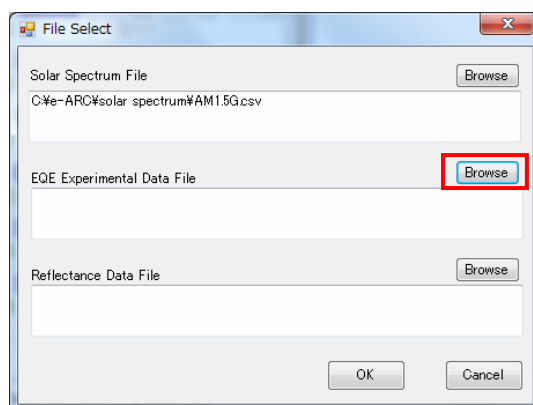
③ In the generated “File Select” subwindow, click “Browse” button of “Solar Spectrum File” (red square below)



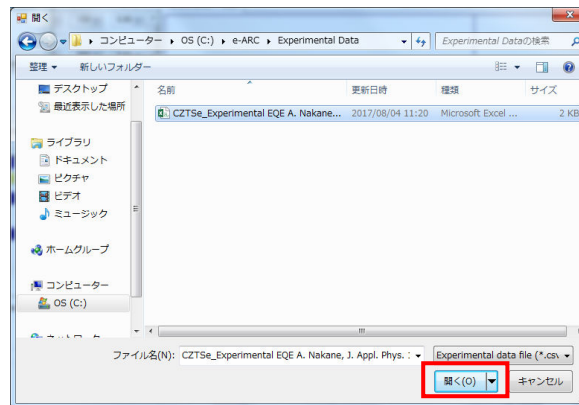
④ Select a file of solar spectrum data (“AM1.5G.csv” in “solar spectrum” directory)



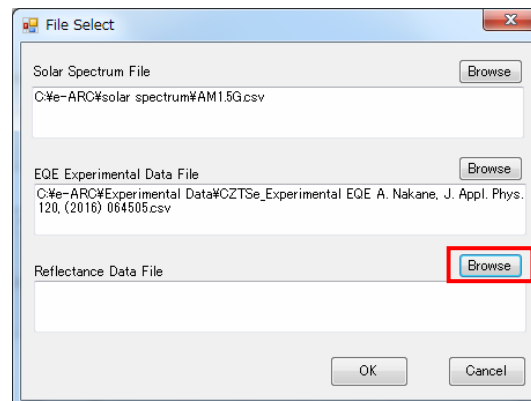
⑤ In the case of the EQE fitting analysis, click “Browse” button of “EQE Experimental Data File”. When only EQE simulations are performed, the selection of EQE is not necessary. The EQE experimental file should have two columns with the first and second columns to be the wavelength and EQE data, respectively.



- ⑥ Select a file of EQE experimental data (“CZTSe\_Experimental EQE A. Nakane, J. Appl. Phys. 120 (2016) 064505.csv” in this example)



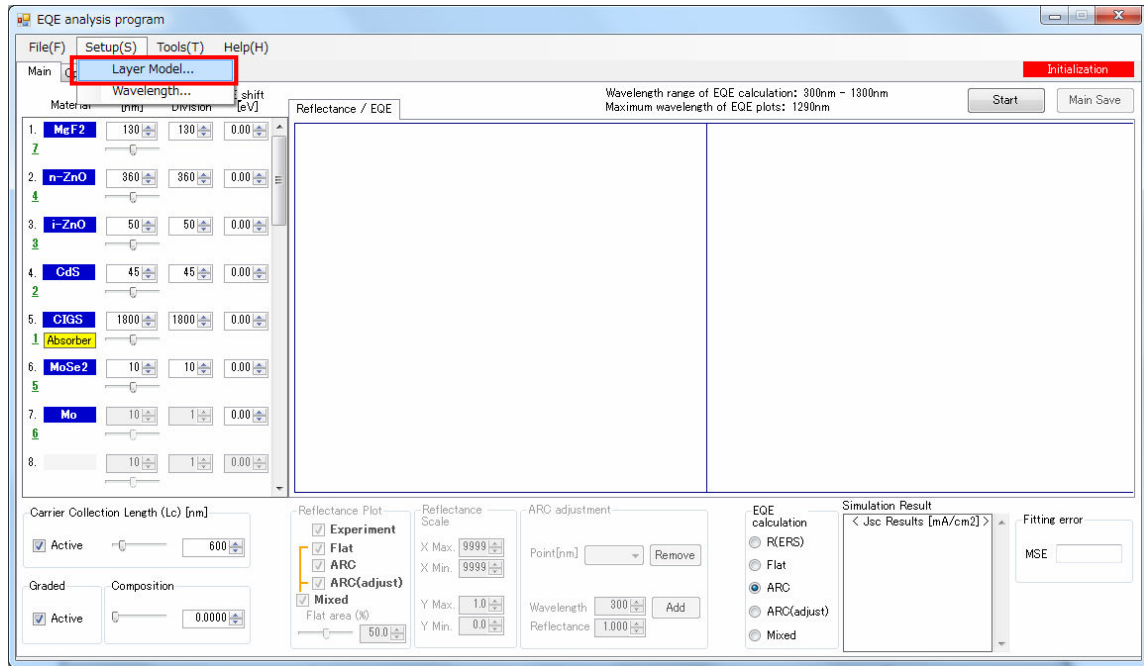
- ⑦ When the experimental  $R$  spectrum is used in the EQE analysis, click “Browse” button of “Reflectance Data File”. The file structure is identical to that of the EQE file mentioned above although the data range could be varied.



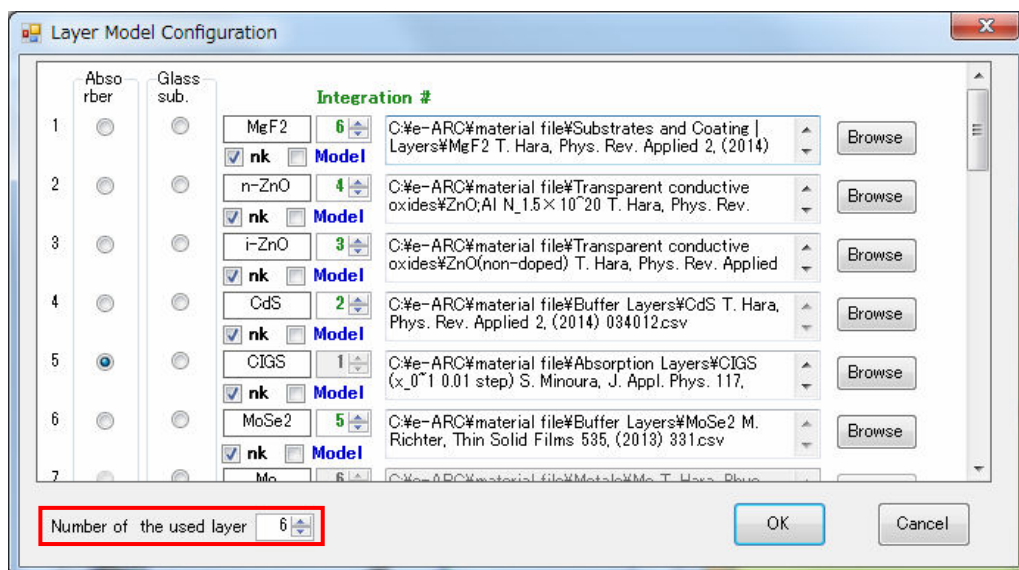
## 4.2 Modeling of a solar cell structure

### 4.2.1 Setting of layer number and layer names

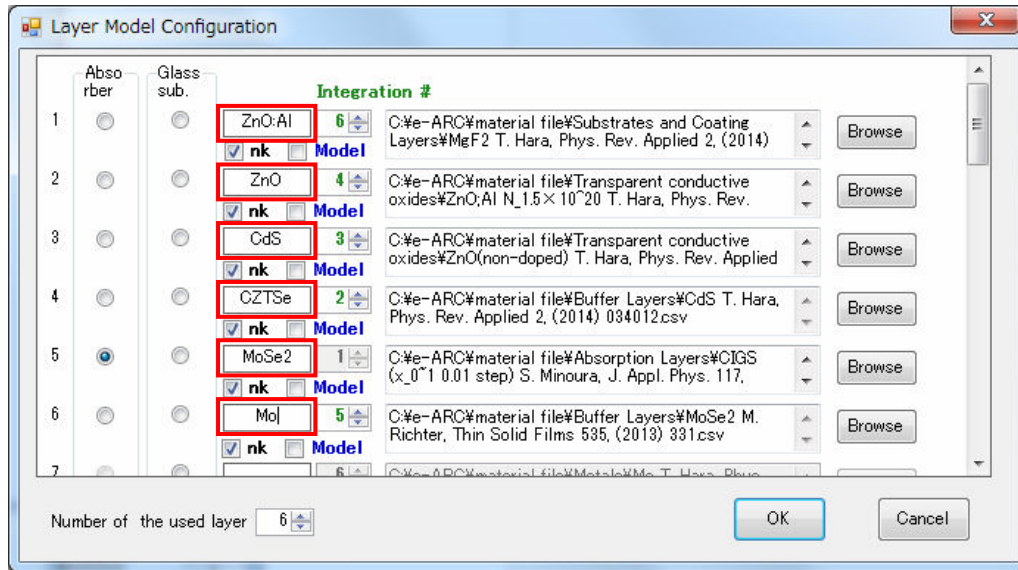
① For the construction of a solar cell structure, click the tab menu of “Setup” and further click “Layer Model”.



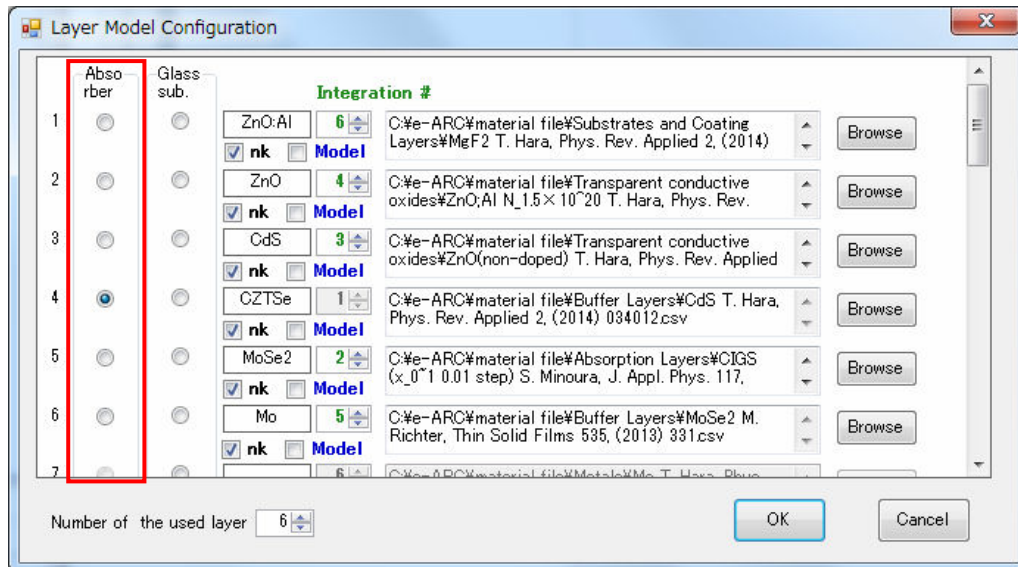
② The CZTSe solar cell has a structure of ZnO:Al/ZnO/CdS/CZTSe/MoSe<sub>2</sub>/Mo. Thus, in the generated subwindow (“Layer Model Configuration”), input “6” for “Number of the used layer” indicated by the red square.



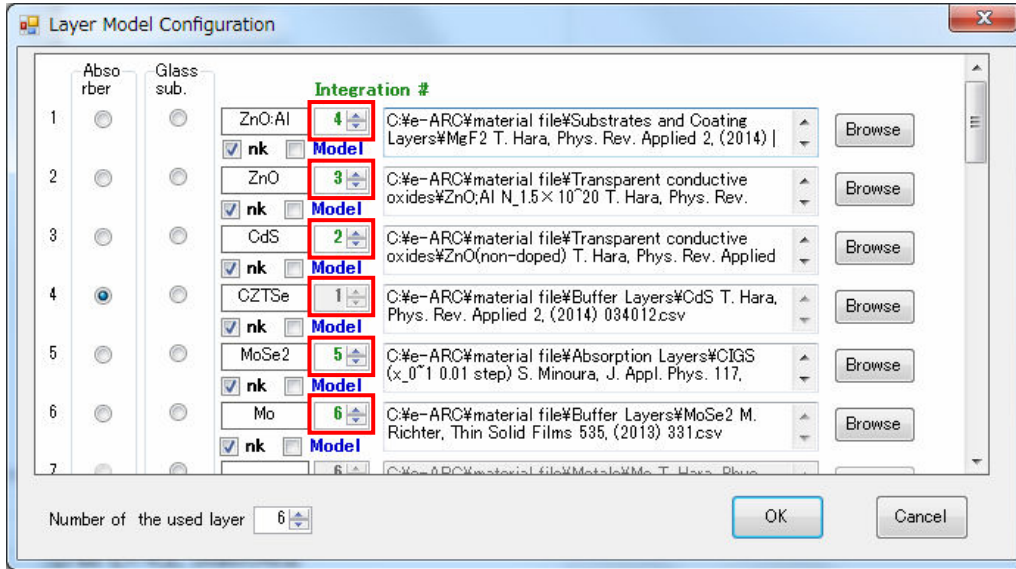
- ③ Input the name of individual layer within the textbox indicated by the red squares.



- ④ For the absorber layer (active semiconductor layer) of the solar cell, click the button of “Absorber” subwindow. In this example, select the CZTSe layer as the absorber. This button needs to be used and affects the color map of the EQE spectrum and  $J_{sc}$  calculation result. The “Glass substrate” subwindow is not used in this example and will be applied for superstrate-type solar cells (see Section 6.1).



⑤ The number of “Integration#” denoted by the green color shows the sequence of the integration in the EQE color-map spectra calculated later. The absorber layer is #1 layer and, in the EQE plot,  $A$  of #2 layer is added to the EQE of #1. When the anti-reflection layer is present, select the number of this layer as the last number. In this example, the integration sequence is as follow: CZTSe(#1), CdS(#2), ZnO (#3), ZnO:Al (#4), MoSe<sub>2</sub> (#5), Mo(#6).



#### 4.2.2 Setting of optical constant data

For the optical constants of layers stored in the “material file” directory (see Section 2.3), two different data types are present: (i) (n, k) data (refractive index n and extinction coefficient k) and (ii) dielectric-function model parameters. The file type of the (n, k) data is “.csv” and this text data has a file structure of Fig. 4.1(a) (i.e., the columns of wavelength, n and k). On the other hand, the file type of the model parameters is “.txt” and the example of the text data is shown in Fig. 4.1(b). In this program, the four dielectric function models are supported: (i) Tauc-Lorentz (TL) model, (ii) Drude model, (iii) Sellmeier model and (iv) Cauchy model (see Refs. [4,5] for the TL model). The sequence of parameter description and the units of each model are summarized in Table 4.1. In the example of Fig. 4.1(b), the CZTSe dielectric function is modeled by a total of five TL peaks [6] and the first row shows the TL parameters of the first TL peak (i.e.,  $A=11.122$  eV,  $C=1.330$  eV,  $E_0=1.177$  eV,  $E_g=0.700$  eV and  $\varepsilon_1(\infty)=0.770$ ).

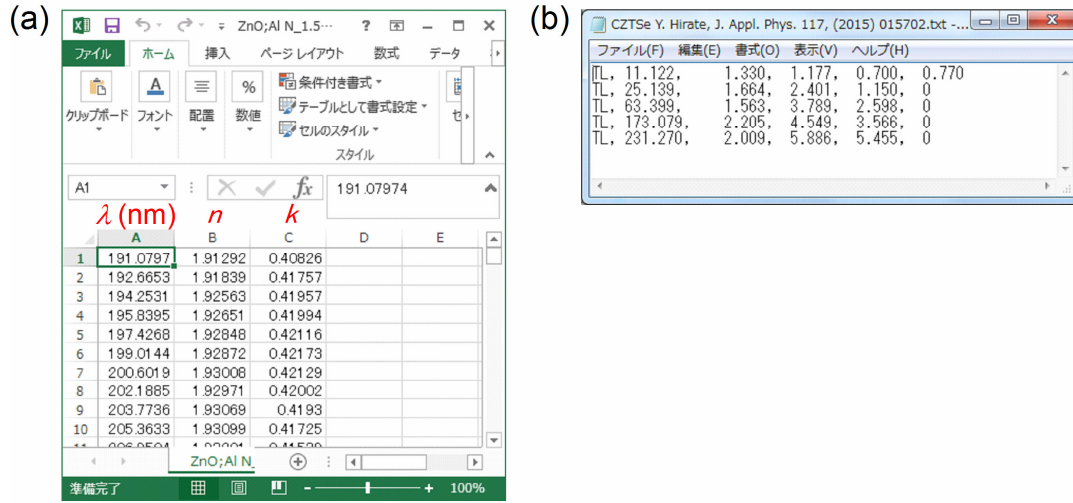


Figure 4.1 (a) ( $n$ ,  $k$ ) data file (“.csv” files) for ZnO:Al and (b) dielectric-function model parameters (“.txt” files) for CZTSe [6].

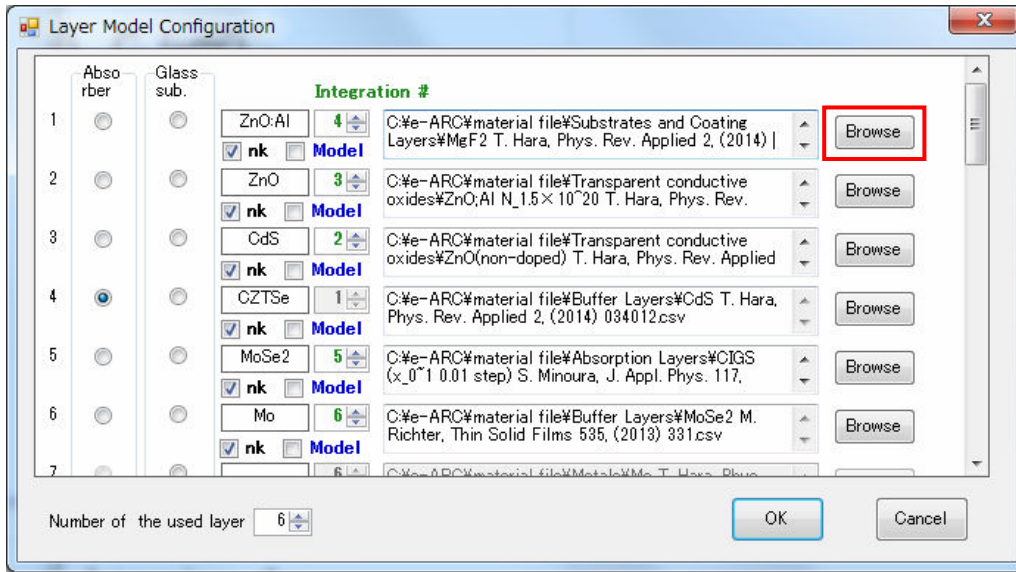
Table 4.1 Sequence of parameter description and units of each model.

Model	Sequence of parameter values					
Tauc-Lorentz(TL)	$A$ (eV)	$C$ (eV)	$E_0$ (eV)	$E_g$ (eV)	$\varepsilon_1(\infty)$	
Drude	$A_D$ (eV)	$\Gamma$ (eV)				
Sellmeier	$B_1$	$B_2$	$B_3$	$C_1$ ( $\mu\text{m}^2$ )	$C_2$ ( $\mu\text{m}^2$ )	$C_3$ ( $\mu\text{m}^2$ )
Cauchy	$A$	$B$ ( $\mu\text{m}^2$ )	$C$ ( $\mu\text{m}^4$ )	$D$ ( $\mu\text{m}^{-2}$ )		

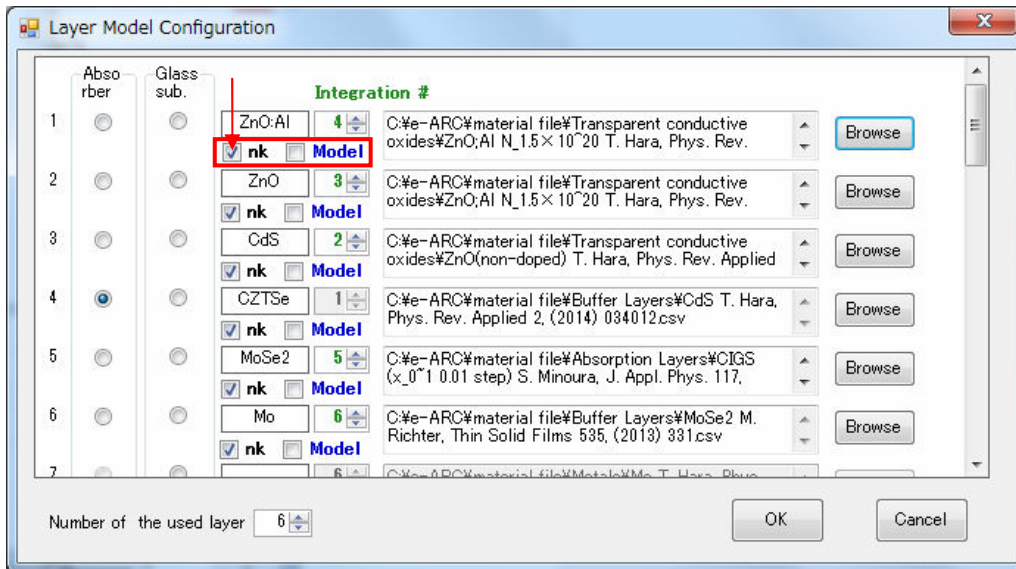
The procedure for the selection of the optical constant data is as follows:

- ① Select the ( $n$ ,  $k$ ) data file for the ZnO:Al using “Browse” button in “Layer Model Configuration” window as indicated by the red square.



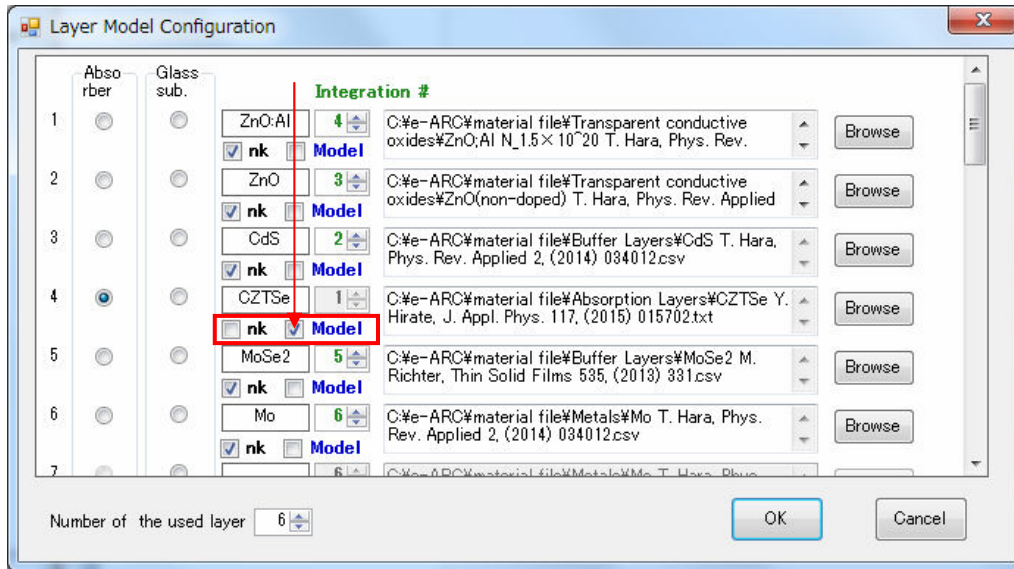


② Select the (n, k) file of “ZnO:Al  $N_{1.5 \times 10^{20}}$  T. Hara, Phys. Rev. Applied 2, (2014) 034012”. The number of this file indicates the optical carrier concentration  $N_{\text{opt}}$  of  $1.5 \times 10^{20} \text{ cm}^{-3}$ , which determines the free carrier absorption in TCO layers [1,4,7]. The reference information is also included as a file name. Then, click the check box of “nk”, as the optical data of the ZnO:Al is in the (n, k) format (see the arrow below).

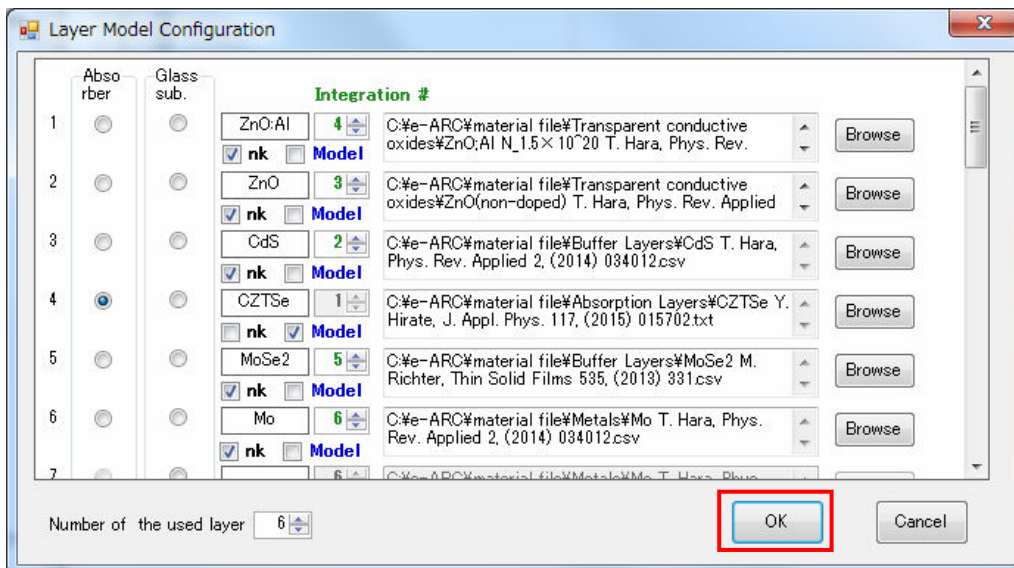




③ Select the input files for all the layers. In this analysis example, the model parameters (i.e, “.txt” file) are used for the CZTSe absorber. Thus, for this layer, click the check box of “Model” indicated by the red arrow.

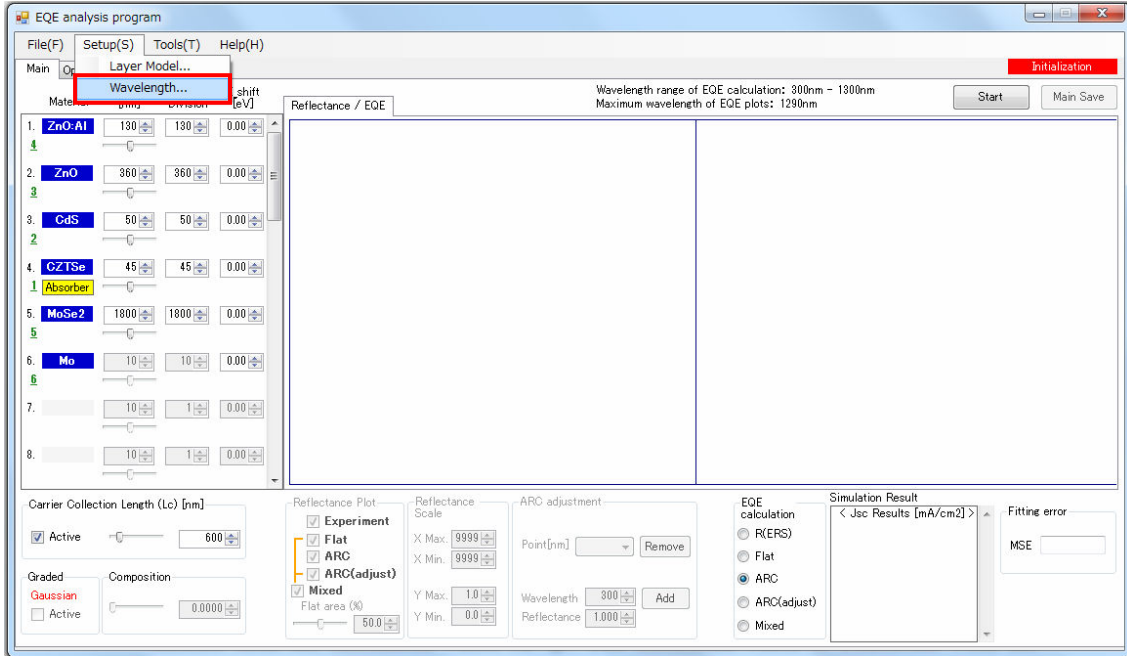


④ The figure below shows the window obtained after finishing all the selection and necessary input. Then, to finish the modeling, click “OK” indicated below.

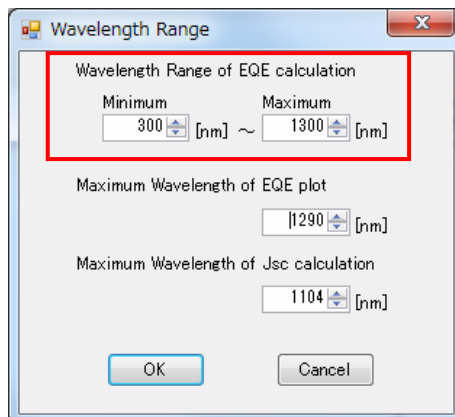


### 4.3 Setting of wavelength range for EQE calculations

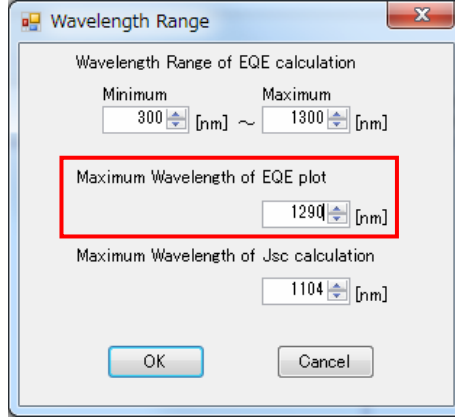
- ① Click the tab menu of “Setup” and further select “Wavelength”



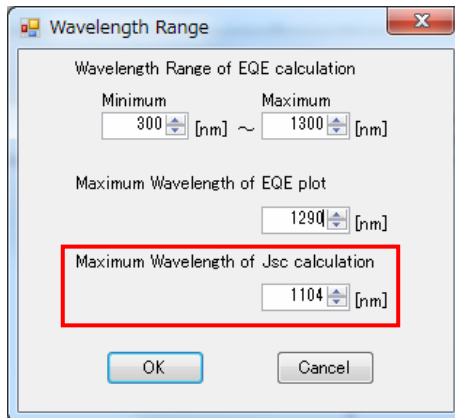
- ② In the generated “Wavelength range” window, insert the minimum and maximum wavelengths as the wavelength range of EQE calculations. In this wavelength range, the data spacing of the experimental data is adjusted automatically to that of the solar light spectrum, so that  $J_{sc}$  can be calculated. It should be emphasized that all the input data (experimental data of EQE,  $R$  and optical constants) need to cover the wavelength range defined by the minimum and maximum wavelengths in this window. The maximum values for “Minimum” and “Maximum” wavelengths are 400 and 3999 nm, respectively.



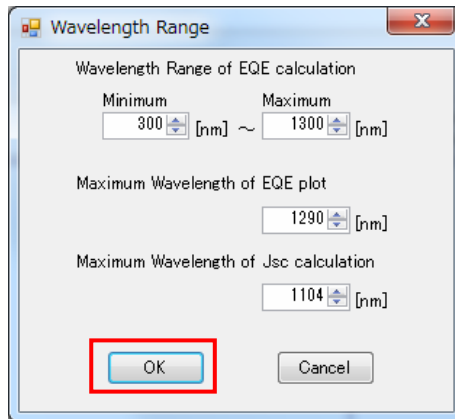
③ Then select the maximum wavelength for the EQE plot. This value should be smaller than the maximum value of ②. The minimum value is consistent with the minimum wavelength range of ②.



④ Input the maximum wavelength for  $J_{sc}$  calculation. This value should be smaller than the maximum value of ②. The minimum value is consistent with the minimum wavelength range of ②. This maximum value is quite important in the analysis of the current loss mechanism as  $J_{sc}$  and other current losses are calculated by integrating the spectra range using this maximum wavelength. Often, solar cells show strong tail absorption and, if the maximum wavelength is set to  $E_g$ ,  $J_{sc}$  is underestimated. On the other hand, if the maximum value is too large, the contribution of the parasitic absorption will be overestimated.



⑤ After setting all the parameters, click “OK”.



#### 4.4 EQE calculation using the main menu.

Figure 4.2 shows the main screen of the software. In this figure, the areas indicated by the red squares represent the windows that control the input parameters (i.e., A~D), whereas the blue squares denote the subwindows that show the calculation results (i.e., E~G).

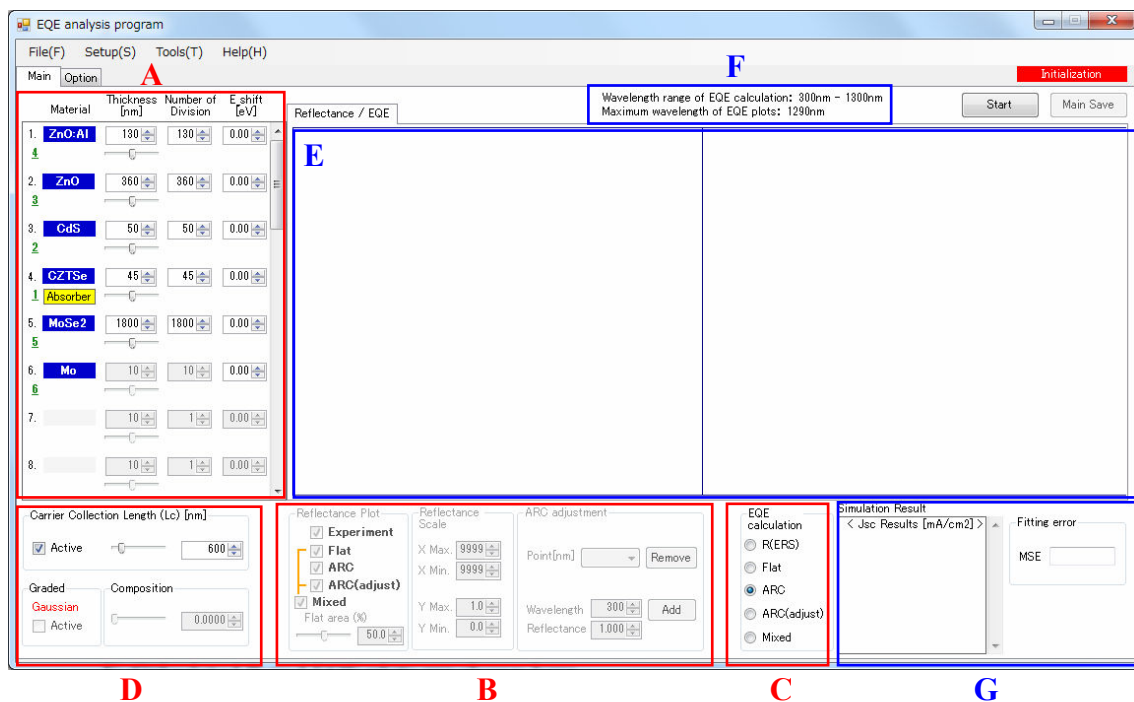


Figure 4.2 Main screen of the e-ARC software.

The details of each panel are described below:

- A: Layer name and thickness, number of division and energy shift value of the optical data.
- B: Detailed setting and adjustment of  $R$  calculation
- C: Selection of the  $R$  spectrum used for the EQE calculation
- D: Recombination analysis (carrier collection length  $L_C$ ) and graded/compositional modulation control.
- E: Calculated  $R$  and EQE spectra are shown here.
- F: This small window indicates the wavelength ranges set at Section 4.3.
- G: Calculation result of  $J_{sc}$ , current losses, and MSE in the EQE fitting analysis.

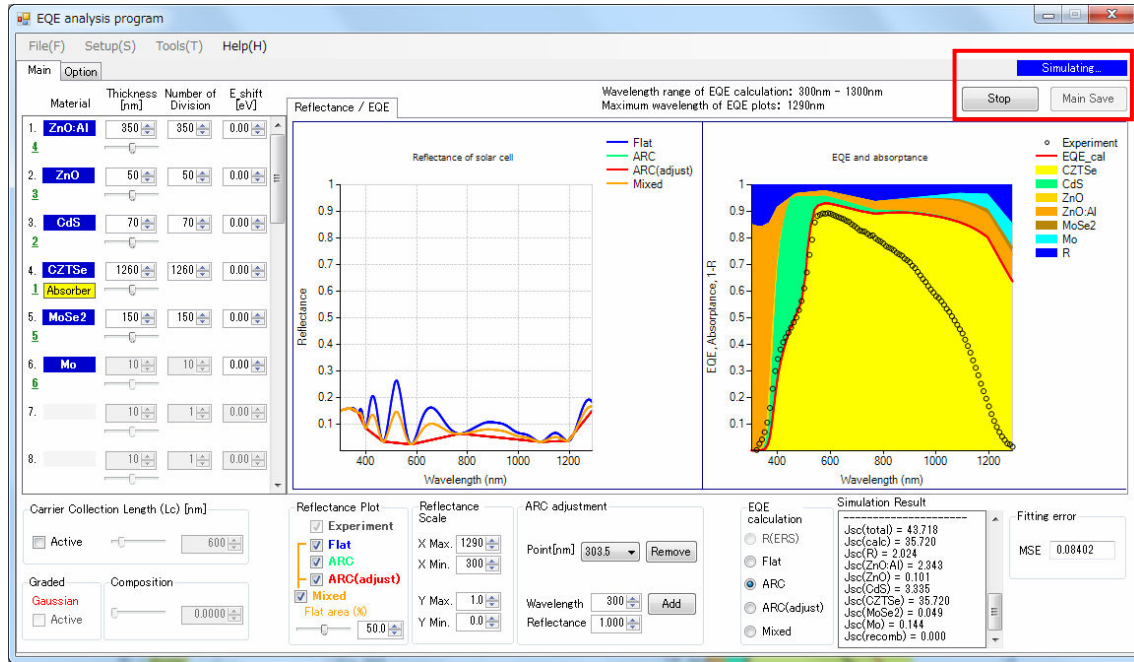
#### ① Setting of solar cell structure (window A)

Input thicknesses of each layer in the text boxes. Set all the numbers of divisions to the same values as the layer thicknesses. The number of division indicates the number of sublayers. If the number of division is 10, this layer is divided into 10 sublayers with an equal thickness. This value becomes important in the depth-resolved EQE calculation (see Option in Section 4.9).

	Material	Thickness [nm]	Number of Division	E_shift [eV]
1.	ZnO:Al	350	350	0.00
2.	ZnO	50	50	0.00
3.	CdS	70	70	0.00
4.	CZTSe	1260	1260	0.00
5.	Absorber	150	150	0.00
6.	MoSe2	150	150	0.00
7.	Mo	10	10	0.00
8.		10	1	0.00

## ② Start EQE calculation

If “Start” button indicated by the red square below is clicked, the EQE calculation starts and the status bar changes to “Simulating”. The “Start” button also changes to “Stop” button. In this status (real-time mode), if the thickness or other parameter is changed, the calculation result is reflected in real time, making the EQE fitting more easily. On the other hand, if “Stop” button is pushed, the status changes to “Idle” and the calculation stops. The calculation result can be saved only during the idle status.



## ③ Setting of the energy shift value (window A)

Often, semiconductor layers show slight  $E_g$  shifts due to the compositional modulation or the Burstein-Moss shift in TCO layers. In the e-ARC software, the effect of  $E_g$  change can be incorporated by simply setting the energy shift value “E\_shift” in the window A. In this case, not only the positive values, but also the negative values can be employed, and the original spectrum is shifted horizontally along the energy direction. At this stage, the feature of the E\_shift can be applied only when the TL model parameters are available and is not supported for the (n, k) data file, although the shifted spectra can be made manually. For all the optical data of Ref. [1], the TL parameterization has been performed and thus the energy shift can be implemented. In the actual calculation, the  $E_0$  and  $E_g$  parameters of the TL model are changed according to the E\_shift value.

In the above analysis example, the energy shift is possible for the CZTSe absorber layer and the value of 0.25 eV is assumed in the analysis. In the calculated result, the

longer wavelength EQE response decreases rather largely due to the spectral shift toward higher energy.

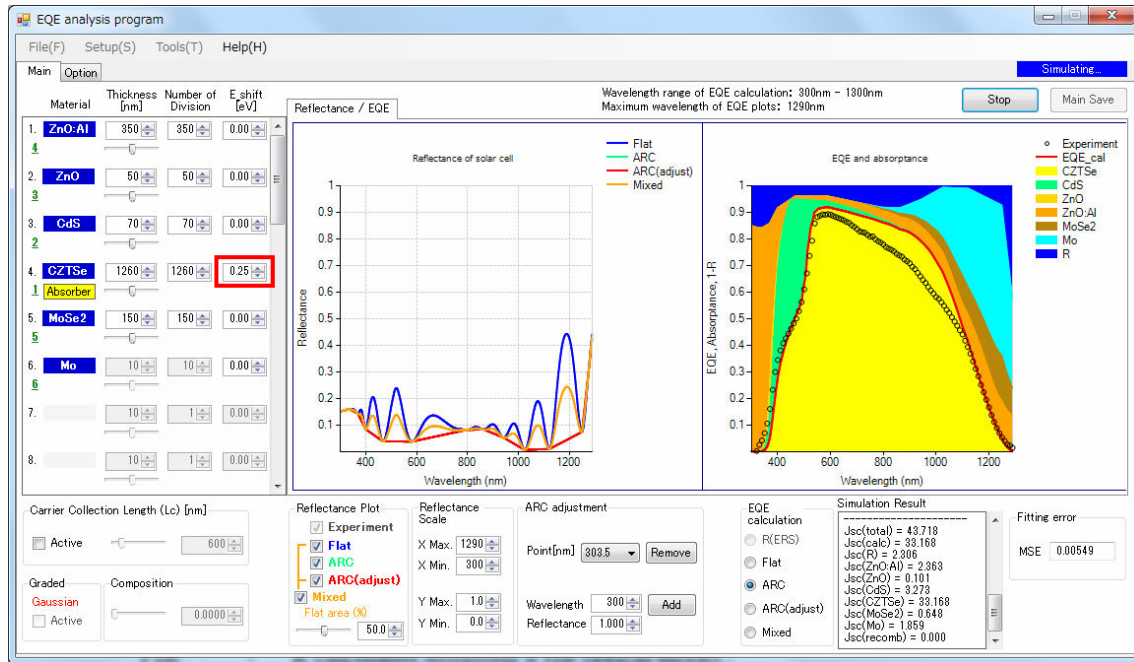


Figure 4.3 EQE analysis result obtained with the energy shift of 0.25 eV in the CZTSe layer.

#### ④ Setting of $R$ spectrum (window B)

Figure 4.4 shows the enlarged window B. In the regions I and II of Fig. 4.4, we select the types and scales of  $R$  spectra, respectively.

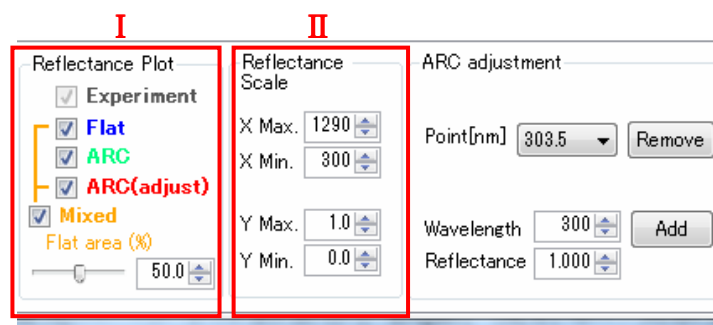


Figure 4.4 Enlarged window B.

In the region I, the  $R$  data for plots are selected. The  $R$  data types are as follows:

Experiment: Experimental  $R$  (this check box appears only when the experimental file is selected).

Flat:  $R$  calculated assuming a flat optical model.

ARC:  $R$  calculated for textured structures using the ARC method [1-3]. This spectrum is obtained by connecting the minimum  $R$  points of the flat structure.

ARC(adjust):  $R$  calculated using the ARC method but with adjusted  $R$  points (see ⑤ below for more detail)

Mixed: In this case,  $R$  is calculated by mixing  $R$  contributions of flat and ARC calculations:

$$Mixed = Flat \times \frac{a}{100} + ARC(adjust) \times \frac{100 - a}{100} \quad (4.1)$$

The value of  $a$  is set as a value of “Flat area (%)” in the window.

In the region II, the scales of the  $R$  plot shown in window E can be changed.

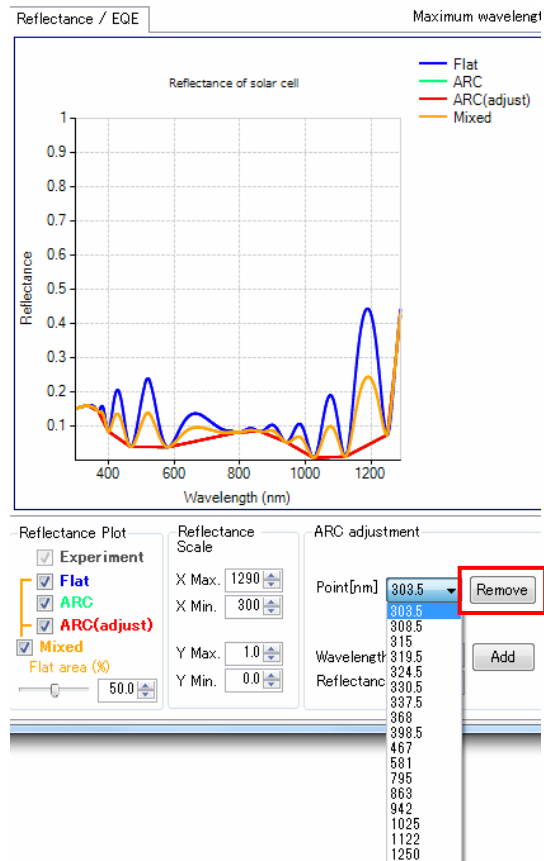
#### ⑤ Adjustment of the $R$ spectrum calculated by the ARC method (window B)

In some cases, the  $R$  spectra obtained from the ARC calculations deviate from experimental  $R$  spectra due to the simplification in the ARC method. In such cases, we can adjust the  $R$  spectra by adding/removing the control  $R$  points using the following procedure.

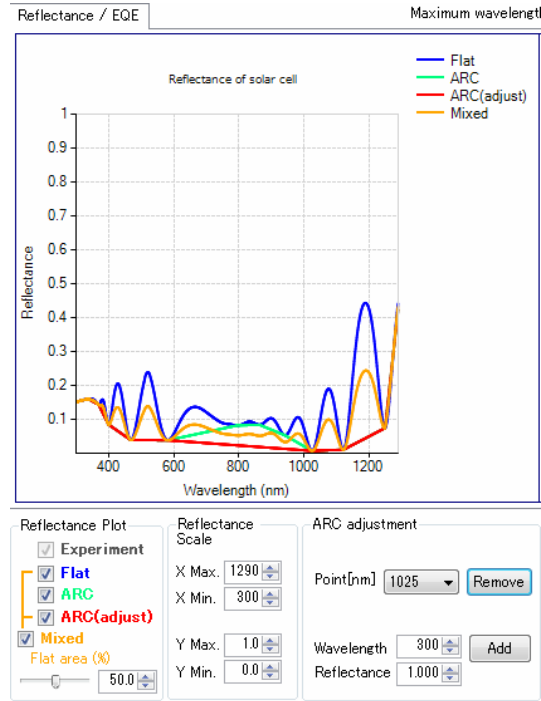
##### I. Removal of the $R$ point

Within the ARC adjustment window, if the numerical value of “Point (nm)” is clicked, the minimum  $R$  points are listed in the column (see below). Then, select a minimum  $R$  point you want to remove.





After selecting the wavelength, click “Remove”, then the result is reflected automatically. In the plot, the modified spectrum is shown as ARC(adjust) as shown below. If necessary, repeat the above procedure to further remove additional  $R$  points.



## II. Addition of the $R$ point

Within the ARC adjustment window, input a  $R$  value at an arbitrary wavelength in the text boxes and then push “Add” button. The added point is shown automatically in the plot.

### ⑥ Selection of the $R$ spectrum for EQE calculation (window C)

In the window C, the  $R$  spectrum used for the EQE calculation is selected. The  $R(ERS)$  in the window indicates  $R$  of experimental reflectance spectrum. Figure 4.5 shows the calculation result when the  $R$  spectrum calculated from the ARC method is chosen. In Figure 4.6, the EQE spectrum obtained when the  $R$  spectrum of the flat model is used is indicated. As confirmed from these two results, the choice of the  $R$  spectrum affects the calculation result largely. In particular, when the texture is present, the interference pattern is eliminated by the light scattering effect and the ARC spectrum needs to be used [2,3].

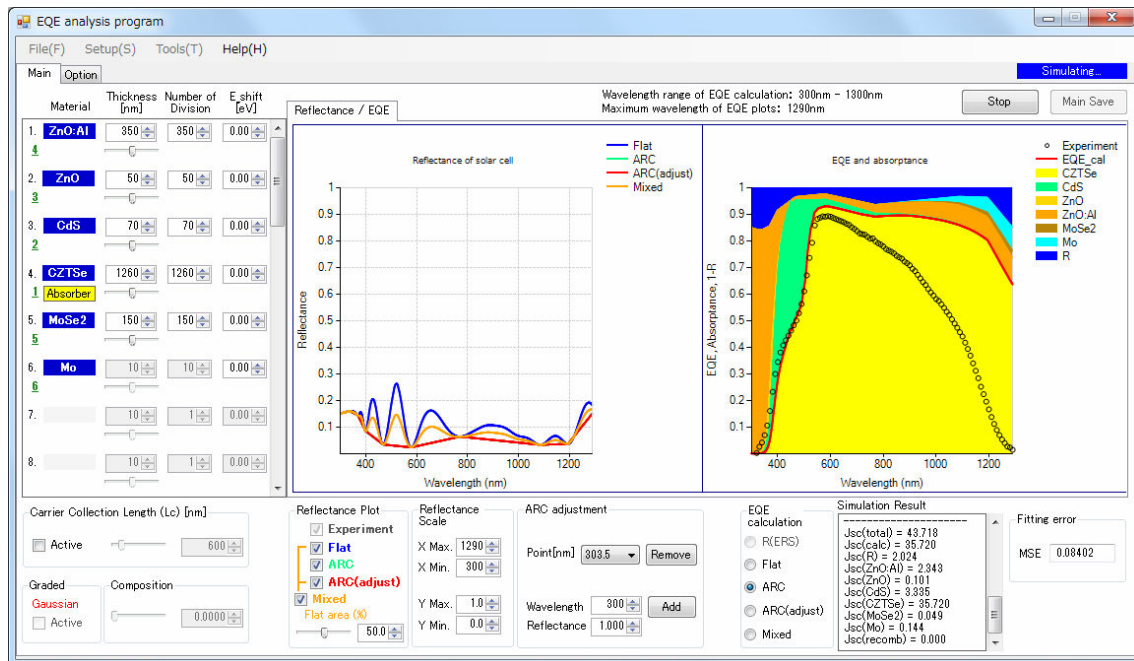


Figure 4.5 Calculation result when the  $R$  spectrum calculated from the ARC method is used.

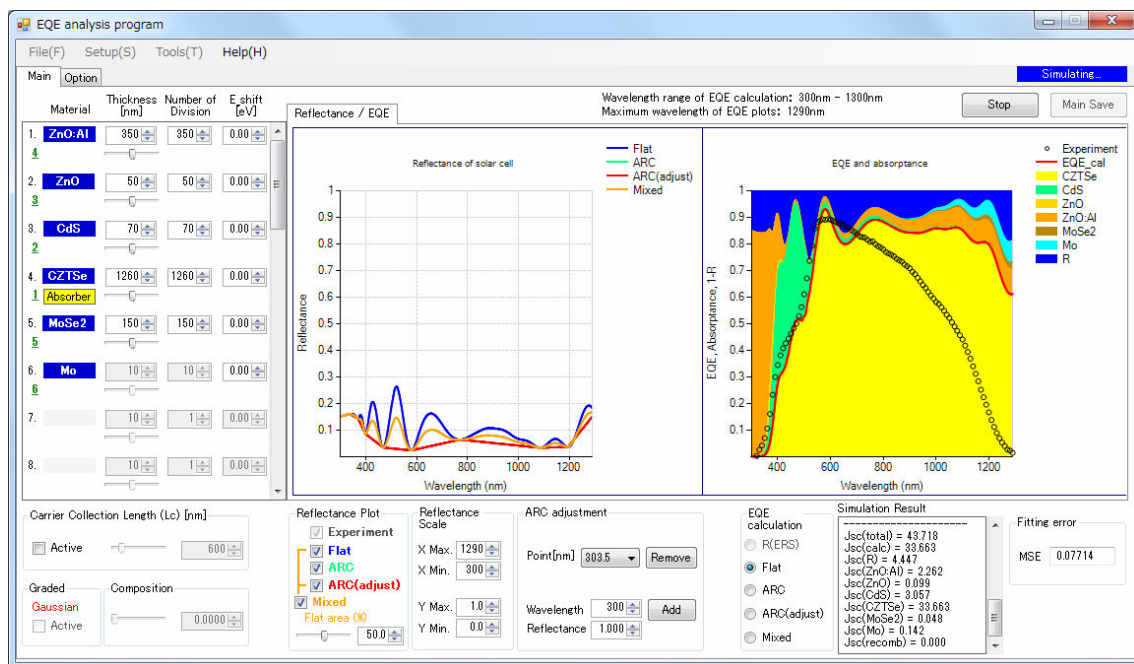


Figure 4.6 Calculation result when the  $R$  spectrum of the flat model is used.

In Figs. 4.5 and 4.6, the red lines in window E (right side) show the calculated EQE spectra, whereas the yellow region indicates the light absorption in the CZTSe absorber layer. In these analysis examples, the agreement of the calculated EQE spectrum with

the experimental result (open circles) is poor. This is caused by the intense carrier recombination in the CZTSe layer (see Section 4.6).

#### 4.5 Calculation result (window G)

After the EQE calculation, the analysis result and MSE are shown in window G (see Fig. 4.7 below). In the “Simulation result” subwindow (region I), the values of the analysis parameters (layer thicknesses, etc.),  $J_{sc}$ , and various current losses are indicated, while the value in the “Fitting error” subwindow indicates the difference between the experimental and calculated EQE spectra.

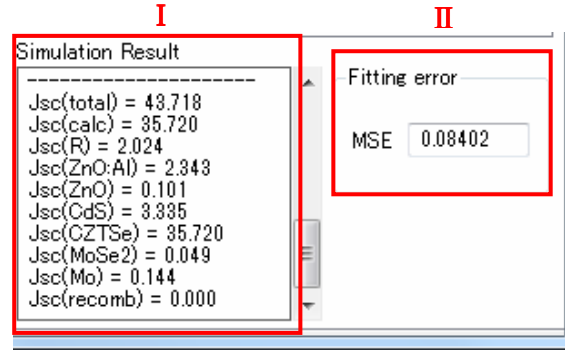


Figure 4.7 EQE analysis result and fitting error (MSE).

In the result subwindow, the date/time, analysis condition and  $J_{sc}$  result are summarized. The various  $J_{sc}$  values are calculated by the following equation [1-3]:

$$J_{sc} = \frac{e\lambda}{2\pi\hbar c} \int F(\lambda)S(\lambda)d\lambda, \quad (4.2)$$

where  $e$  and  $c$  show the electron charge and speed of light, respectively. In the above equation,  $F(\lambda)$  is the spectrum or a function described below, while  $S(\lambda)$  indicates the solar light spectrum (irradiance). The integration of Eq. (4.2) is carried out in the range selected in Section 4.3.

Depending on the selection of  $F(\lambda)$ , different  $J_{sc}$  values are calculated as listed below:

Jsc(total):  $J_{sc}$  obtained by setting  $F(\lambda)=1$  in Eq. (4.2). This corresponds to the maximum  $J_{sc}$  possible in the selected wavelength range (i.e., all the photons are simply converted into the current).

Jsc(calc):  $J_{sc}$  obtained by setting  $F(\lambda)=EQE(\lambda)$ . This  $J_{sc}$  is consistent with the calculated EQE spectrum. If the fitting to the experimental EQE spectrum is sufficiently good, the Jsc(calc) should show a similar value close to the experimental  $J_{sc}$ .

- Jsc(R):  $J_{sc}$  obtained by setting  $F(\lambda)=R(\lambda)$ . This shows the  $J_{sc}$  loss induced by the light reflection. Jsc(R) is calculated by  $R(\lambda)$  selected in the window C.
- Jsc(layer):  $J_{sc}$  obtained by setting  $F(\lambda)=A(\lambda)$ . This shows the  $J_{sc}$  loss induced by the unfavorable parasitic light absorption in each solar-cell component layer.
- Jsc(recomb):  $J_{sc}$  obtained by setting  $F=A(\text{absorber})-EQE$ . This shows the  $J_{sc}$  loss induced by the carrier recombination. If  $J_{sc}$  of the absorber layer is Jsc(absorber), Jsc(recomb) is expressed as Jsc(recomb)=Jsc(absorber)-Jsc(calc).

#### 4.6 Analysis of carrier recombination (window D)

In the e-ARC method, the carrier recombination that occurs within the absorber layer can further be analyzed [1-2]. The carrier recombination can be modeled rather simply by defining the carrier collection length  $L_C$ . As illustrated in Fig. 4.8,  $L_C$  indicates a length from the absorber front interface and the carrier generated within the distance of  $L_C$  is collected while the other carriers generated in the solar cell bottom region is lost by the recombination. In the e-ARC method, the carrier collection efficiency  $H(\lambda)$  is modeled as

$$H(\lambda) = 1 - \exp[-\alpha(\lambda)L_C], \quad (4.3)$$

where  $\alpha(\lambda)$  shows the absorption coefficient of the absorber (semiconductor) layer. By employing the above equation, the resulting EQE spectrum can be calculated by

$$EQE(\lambda) = A(\lambda)H(\lambda). \quad (4.4)$$

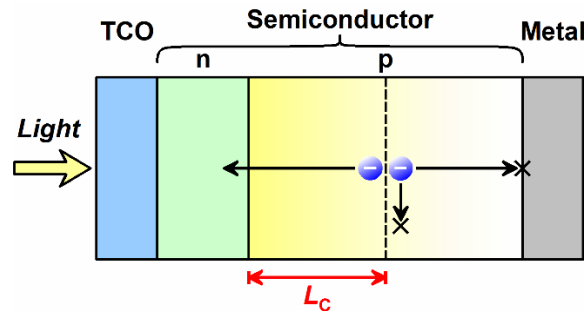


Figure 4.8 Definition of carrier collection length  $L_C$ .

By using the above e-ARC method, the CZTSe analysis in the previous section is

performed again here. To perform the recombination analysis, click “Active” check box in the window D and input the  $L_C$  value (nm) in the text box. The EQE spectrum is updated automatically according to the  $L_C$  value. Figure 4.9 shows the final result obtained from this analysis. In this analysis,  $L_C = 700$  nm and E shift (CZTSe) of 0.15 eV are assumed. From such analyses, all the current loss mechanism (reflection, parasitic absorption and recombination losses) can be determined.

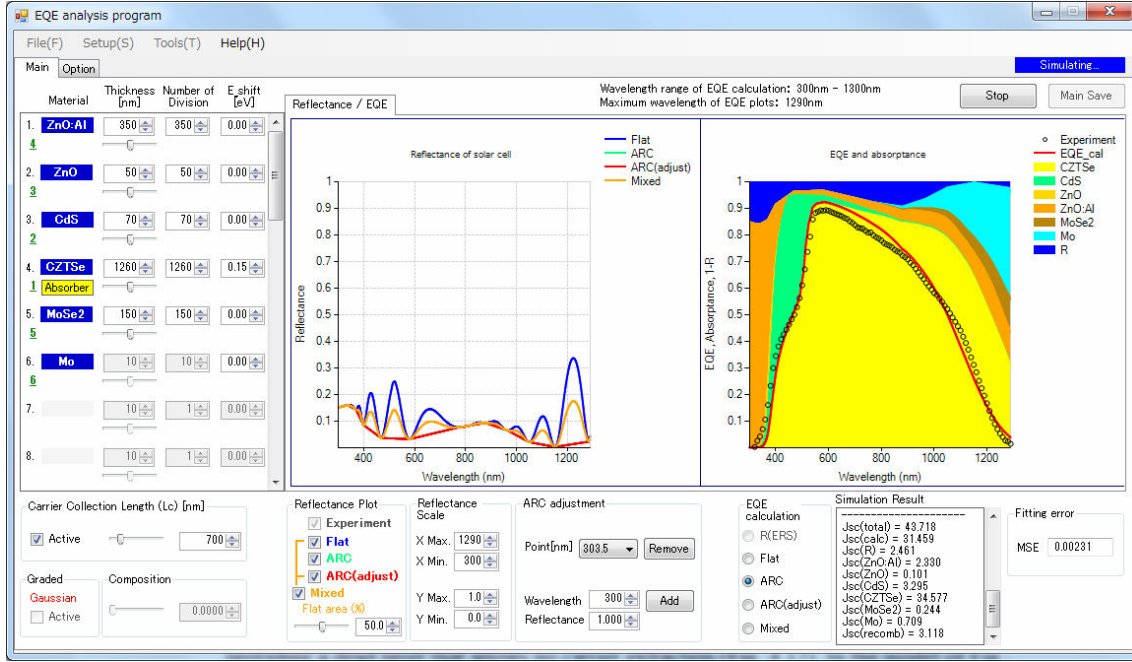


Figure 4.9 Final analysis result obtained for the CZTSe solar cell using the e-ARC method.

In the actual experimental CZTSe solar cell used in the above analysis, rather extensive void formation occurs at CZTSe/Mo interface [2] but this effect is neglected in this example. If more detailed analysis is performed, the EQE fitting improves notably [2].

#### 4.7 Analysis of interface recombination (window A)

In the e-ARC method, the carrier recombination that occurs in the rear interface region can be expressed quite well, but the recombination in the front interface region cannot be described. However, the front interface recombination can be modeled by providing a dead layer that allows no carrier extraction (Fig. 4.10). In the model of Fig. 4.10, the dead layer is placed within the absorber layer with a thickness of  $d_{\text{dead}}$ . If the front interface recombination is strong, the effective thickness of  $d_{\text{dead}}$  increases and the

carrier loss increases.

To perform the above analysis, provide a dead layer at the interface using the same optical constants as the absorber in window A. In other words, in this analysis, we introduce identical two absorber layers in the optical model and treat the one layer as the dead layer while assuming 100% carrier collection in the rest of the absorber layer.

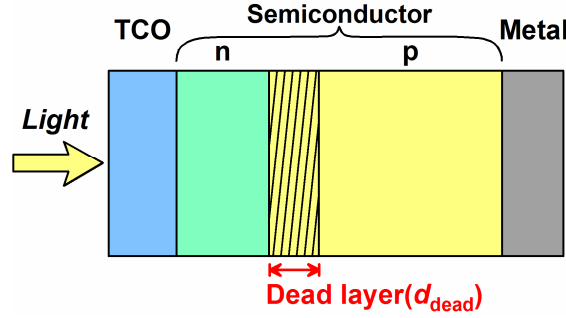


Figure 4.10 Definition of a dead layer in an absorber layer.

The example of the dead layer analysis is shown in Fig. 4.11. In this case, the front interface recombination is considered and the optical model of ZnO:Al/ZnO/CdS/Dead layer (CZTSe)/Absorber (CZTSe)/MoSe<sub>2</sub>/Mo (7 layers) is constructed. In Fig. 4.11, the thickness of the dead layer is 30 nm and, since EQE is calculated from the contribution of only the CZTSe absorber layer, the EQE decreases by the light absorption in the dead layer. In the actual fitting analysis, a total thickness (absorber+dead layers) needs to be maintained at a constant value. As confirmed from Fig. 4.11, the front interface recombination reduces the EQE response in the short wavelength region, while the recombination in the rear interface region decreases EQE in the longer wavelength region. Accordingly, this analysis confirms that the front interface recombination is negligible in the CZTSe solar cell [2].

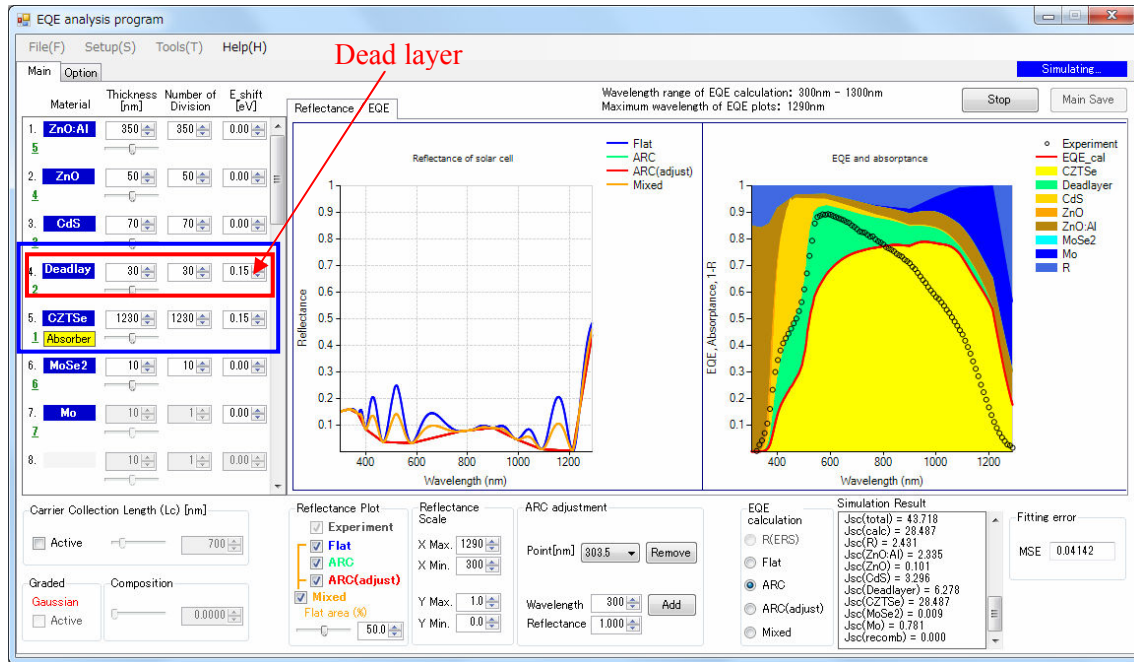


Figure 4.11 Example of the dead layer analysis. The blue square indicates the CZTSe layers and the red square represents the CZTSe dead layer.

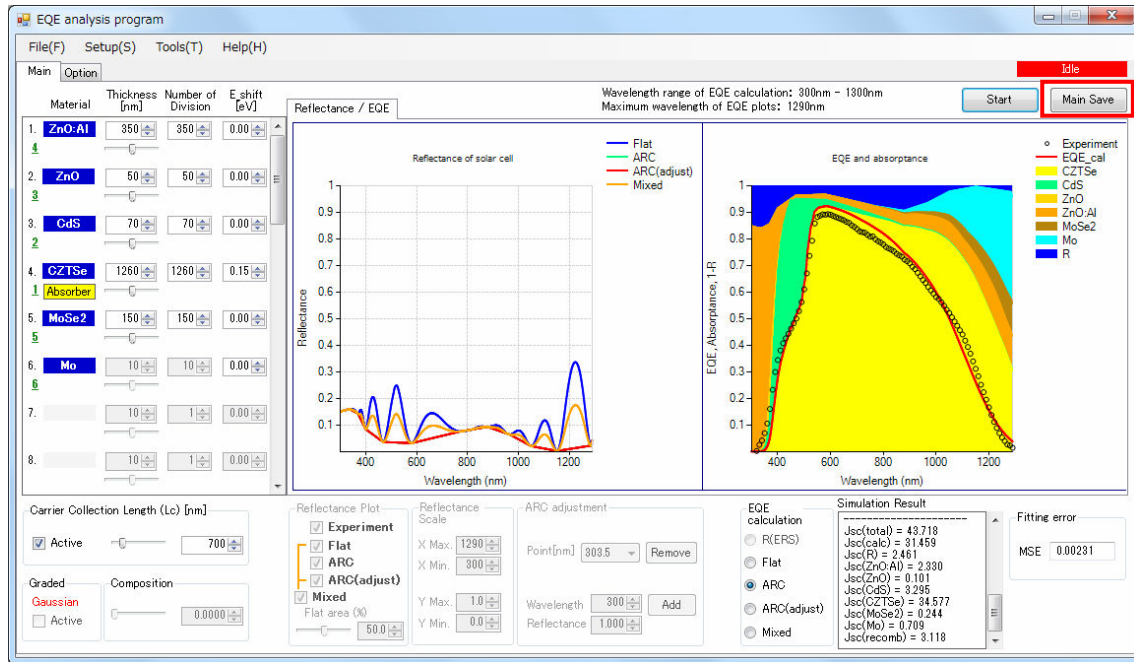
By assuming the rear interface recombination, a more complex analysis can be performed. If the dead layers are assumed to exist at both interfaces (i.e., front and rear interfaces), the absorber layer should be divided into a total of three layers (i.e., front dead layer/absorber/rear dead layer). This analysis can be implemented by further introducing the additional layer in window A. Nevertheless, the combination of the front dead layer with  $L_C$  provides better EQE fitting [1,2].

#### 4.8 Saving the calculation result

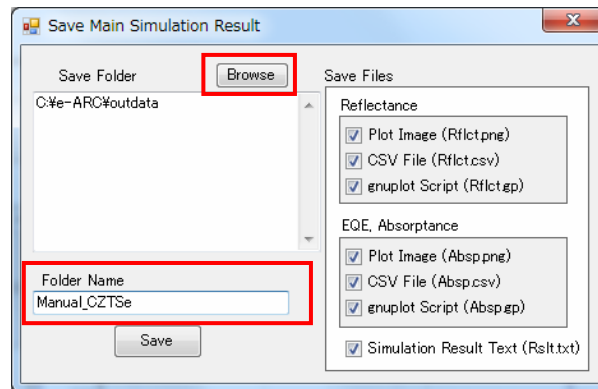
All the calculation results including (i)  $R$  spectra, (ii) EQE and  $A$  spectra, and (iii) simulation results can be saved. To do this,

- ① First click “Stop” button in the main menu to finish the real-time mode. And then click “Main Save” button in the main window (see below).





② In the generated “Save Main Simulation Result” window, click “Browse” button to select the directory and enter the folder name. For the intended files to be saved, click the check boxes. Finally, click “Save” button to finish the operation.



#### 4.9 Optional calculation

In the option window, the depth-resolved EQE and  $A$  spectra (partial EQE and  $A$ ) are calculated. For this, click the “Option” tab located right next to the main tab (near “Setup” menu bar). By this procedure, the option window (see below) is generated:

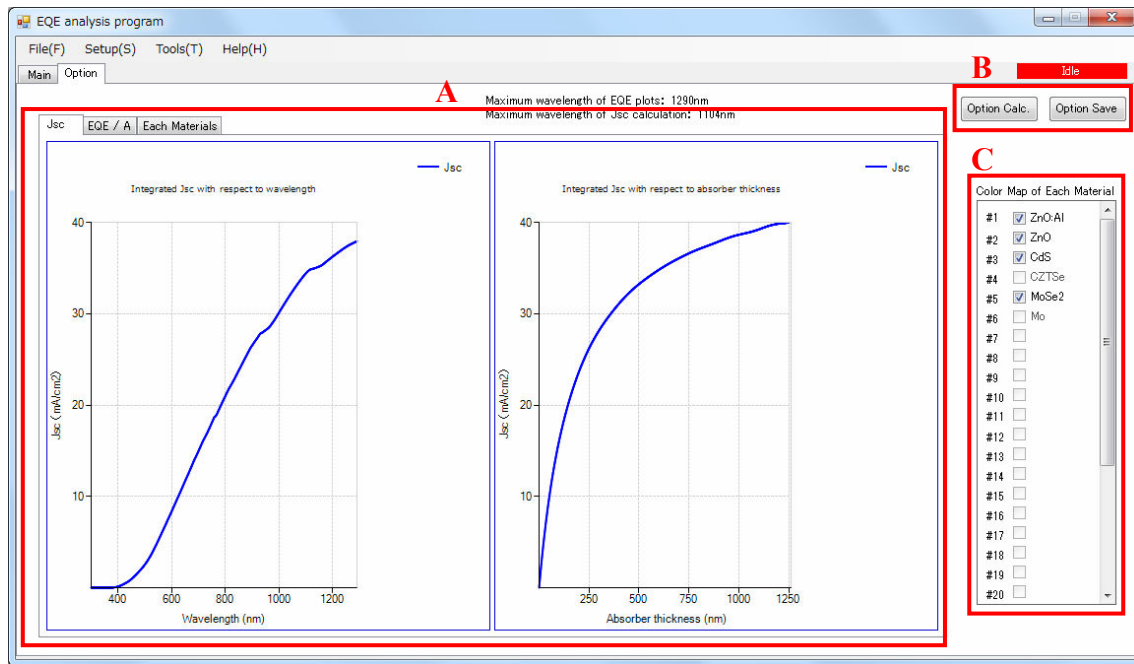


Figure 4.12 Screen of the option window

The windows of the option window are as follows:

A: Result window. There are also sub-menus described below:

Jsc: Integrated  $J_{sc}$  for wavelength and layer thickness is shown. The integrated  $J_{sc}$  is consistent with  $J_{sc}$  of the EQE spectrum.

EQE/A: Depth-resolved color mapping graphs for EQE and  $A$  of the absorber layer. If these partial EQE and  $A$  spectra are integrated, the EQE and  $A$  spectra of the absorber layer in the main window are reproduced.

Each materials: Depth-resolved color mapping graphs for  $A$  of the component layers.

B: Option calculation/save window.

Option calculation and data saving are controlled.

C: Data save window

When the check boxes are clicked, the data of the clicked materials are saved.

#### ① Color mapping data

The following example (Fig. 4.13) shows the partial  $A$  spectra of the ZnO layer in the CZTSe solar cell. To show this data, click “Each material” tab in the window A, then click the “#2” button that appears at the top part of the subwindow. In the solar cell

structure, the second layer on the light incident side is the ZnO layer. Thus, #2 corresponds to the ZnO layer. The setting for the ZnO layer in the main menu (window A) is as follow: thickness = 50 nm, number of division = 50. In this case, since the ZnO layer is divided into a total of 50 layers, the thickness of one sublayer is 1 nm. If the division is 100, this thickness decreases to 0.5 nm. In the mapping data, the partial  $A$  spectra calculated using the corresponding number of division are shown. In the above example, the number of division is set to 1-nm resolution as the thickness is equal to the number of divisions. The depth resolution can be improved by simply increasing the number of divisions.

In the example of Fig. 4.13, the number of division is 2000. As shown in this figure, the light absorption in the ZnO layer occurs only in a selected region. The lower-wavelength limit is determined by the light absorption in the upper ZnO:Al layer, whereas the maximum limit is determined by  $E_g$  of the ZnO layer (Note that  $E_g$  of non-doped ZnO is slightly smaller than that of ZnO:Al due to the Burstein-Moss shift).

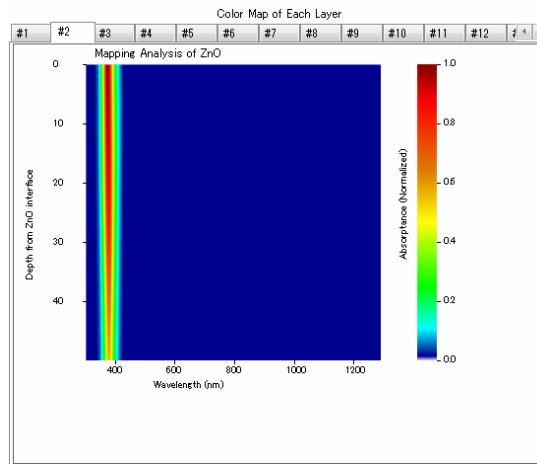
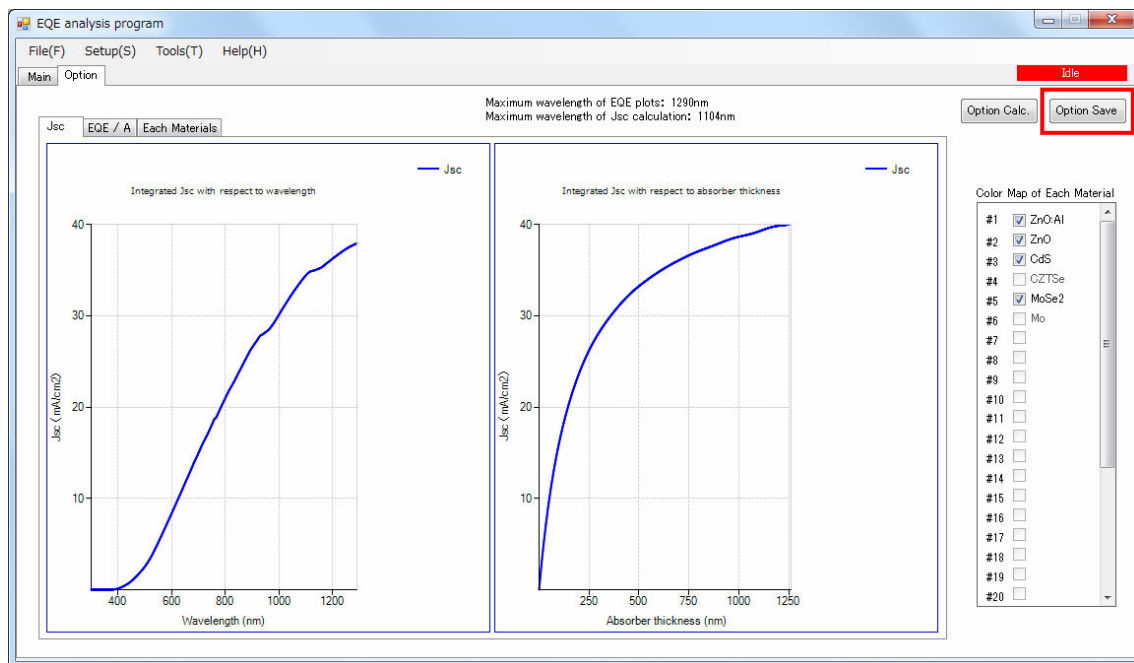


Figure 4.13 Partial  $A$  spectra of the ZnO layer with a thickness of 50 nm (Number of division is 2000).

## ② Saving of color mapping data

First click “Stop” button in the main menu. The calculation results can be saved by clicking “Option Save” in window B (see below). In the generated “Save Option Result” window, the same procedure used in the “Main save” can be used (see Section 4.8).



## 5. Example 2: EQE analysis of a CIGSe solar cell with a double grading structure

In this section, a more complicated EQE analysis for a state-of-the-art CIGSe solar cell fabricated by three-stage coevaporation is described. The experiment and EQE analysis are described in Ref. [3]. The CIGSe solar cell analyzed here has a structure of  $\text{MgF}_2/\text{ZnO:Al}/\text{ZnO}/\text{CdS}/\text{CIGSe}/\text{MoSe}_2/\text{Mo}$  (a total of seven layers).

### ① Modeling of Ga grading structures

For the analysis of the Ga grading structures, the complete (n, k) optical database established for different Ga composition x [i.e.,  $x=\text{Ga}/(\text{In}+\text{Ga})$ ] is necessary. In this example, the CIGSe optical database reported by Minoura et al. [8] are used. To perform the CIGSe analysis, the CIGSe optical-constant data file has already been made and is included in the software package (File: CIGS (x\_0~1 0.01 step) S. Minoura, J. Appl. Phys. 117, (2015) 195703.csv).

Figure 5.1 shows the data structure of this file. In the file, the (n, k) data of  $\text{CuIn}_{1-x}\text{Ga}_x\text{Se}_2$  with a compositional step of 1 at.% (i.e.,  $x=0.01$ ) are summarized. The leftmost column shows the wavelength, and the second and third columns correspond to the n and k data of the  $\text{CuInSe}_2$  ( $x=0.0$ ), while the fourth and fifth columns show the n and k of the CIGSe ( $x=0.01$ ). There are roughly 200 columns to cover the compositional range of  $x=0.00$ -1.00.

Increase in Ga composition x → 1

	0			0.01											
	$\lambda$ (nm)	n	k	n	k										
	A	B	C	D	E	F	G	H	I	J	K	L	M		
1	280.	2.872784	1.248507	2.871747	1.245352	2.870667	1.242229	2.869545	1.23914	2.868379	1.236086	2.867173	1.233066		
2	280.5	2.872093	1.242156	2.870989	1.239058	2.869844	1.235995	2.868656	1.232966	2.867428	1.229974	2.866159	1.227019		
3	281	2.871392	1.235819	2.870221	1.232779	2.86901	1.229774	2.867758	1.226806	2.866466	1.223876	2.865136	1.220985		
4	281.5	2.870701	1.229474	2.869463	1.226491	2.868185	1.223546	2.866869	1.220639	2.865514	1.217771	2.864122	1.214943		
5	282	2.869764	1.223432	2.868466	1.220512	2.86713	1.21763	2.865755	1.214788	2.864345	1.211986	2.862998	1.209226		
6	282.5	2.868759	1.217478	2.867402	1.214621	2.866008	1.211804	2.864579	1.209028	2.863114	1.206294	2.861614	1.203606		
7	283	2.867762	1.211515	2.866346	1.208722	2.864895	1.20597	2.86341	1.203261	2.86189	1.200594	2.860338	1.197971		
8	283.5	2.866613	1.205792	2.865144	1.203064	2.863641	1.20038	2.862105	1.197739	2.860536	1.195142	2.858937	1.19259		
9	284	2.865357	1.200233	2.863838	1.197574	2.862287	1.194959	2.860704	1.192388	2.859091	1.189862	2.857448	1.187382		
10	284.5	2.864108	1.194667	2.862539	1.192076	2.86094	1.18953	2.85931	1.187029	2.857651	1.184574	2.855964	1.182167		
11	285	2.862778	1.189266	2.861162	1.186745	2.859518	1.184269	2.857845	1.181839	2.856145	1.179456	2.854418	1.177121		
12	285.5	2.861324	1.18411	2.859668	1.181659	2.857984	1.179255	2.856273	1.176898	2.854537	1.174588	2.852775	1.172327		
13	286	2.859875	1.178946	2.858178	1.176566	2.856454	1.174234	2.854706	1.171949	2.852933	1.169713	2.851137	1.167526		
14	286.5	2.858394	1.17387	2.856658	1.171562	2.854897	1.169302	2.853112	1.16709	2.851305	1.164928	2.849476	1.162815		
15	287	2.856791	1.169114	2.855022	1.166879	2.85323	1.164682	2.851417	1.162554	2.849582	1.160466	2.847727	1.158427		
16	287.5	2.855191	1.164352	2.85339	1.162189	2.851568	1.160075	2.849725	1.158011	2.847862	1.155997	2.845981	1.154033		
17	288	2.853591	1.159601	2.851757	1.157512	2.849904	1.155471	2.848032	1.153481	2.846142	1.151541	2.844235	1.149651		
18	288.5	2.851883	1.155241	2.850025	1.153224	2.848149	1.151257	2.846257	1.149341	2.844347	1.147474	2.842423	1.145659		
19	289	2.850177	1.150874	2.848296	1.14893	2.846398	1.147037	2.844484	1.145194	2.842555	1.143402	2.840613	1.14166		
20	289.5	2.848476	1.1465	2.84657	1.14463	2.844649	1.142811	2.842714	1.141041	2.840765	1.139323	2.838805	1.137656		
21	290	2.846715	1.142467	2.844792	1.14067	2.842855	1.138923	2.840906	1.137226	2.838945	1.135581	2.836973	1.133986		

Figure 5.1 Data of "CIGS (x\_0~1 0.01 step) S. Minoura, J. Appl. Phys. 117, (2015) 195703.csv".

Similar optical-constant data file can also be established for other alloy materials. In this case, the material file can be made using arbitrary number of alloy compositions. For example, when there are (n, k) files of six different compositions for  $A_xB_{1-x}$  alloys with a step of 0.2, there should be a total of 13 columns in the file including the wavelength column. In this case, the first one pair of the (n, k) data is assigned to the composition of 0.0 automatically and the composition of the sixth pair of the data becomes  $x=1.0$ .

If such an optical database file with different compositions is made, the composition of the absorber can be controlled using the composition window (Window D in Fig. 4.2), as shown below. If there are six compositions, the value can be changed with a step of 0.2. In the case of CIGSe,  $x$  can be changed with a step of  $x=0.01$ .

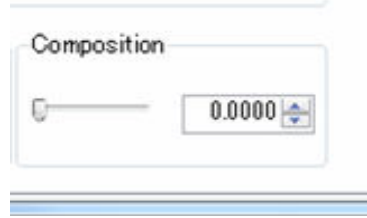


Figure 5.2. Enlarged screen of the composition subwindow.

## ② Modeling of the graded structure

For the analyses of compositional grading structures, the software provides two different models: (i) linear variation model and (ii) double Gaussian model. In the linear model, the composition changes linearly between arbitrary data points. When the double Gaussian model is used, the double Ga grading structures in CIGSe layers are approximated by using a total of six parameters [3]:

$$x = \begin{cases} x_1 - (x_1 - x_v) \exp\{-(d - d_v)/w_1\}^2 & (d \leq d_v) \\ x_2 - (x_2 - x_v) \exp\{-(d - d_v)/w_2\}^2 & (d > d_v) \end{cases} \dots\dots\dots (5.1)$$

where  $d$  is a position and  $d_v$  shows a position of the compositional valley as indicated in Fig. 5.3. The  $x_1$  and  $x_2$  indicate the compositions at the front and rear interface, respectively, whereas  $x_v$  is  $x$  at the valley position. In addition,  $w_1$  and  $w_2$  represent the widths of the first and second Gaussian functions. Note that the values of  $d$  and  $x$  are in the range of 0~1. In particular, the  $d$  values are normalized and the absolute thickness of this layer is set at window A of the main menu (Fig. 4.2).

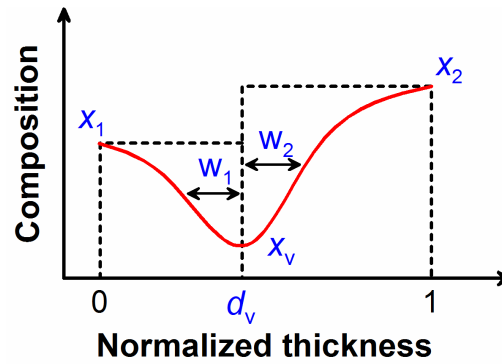
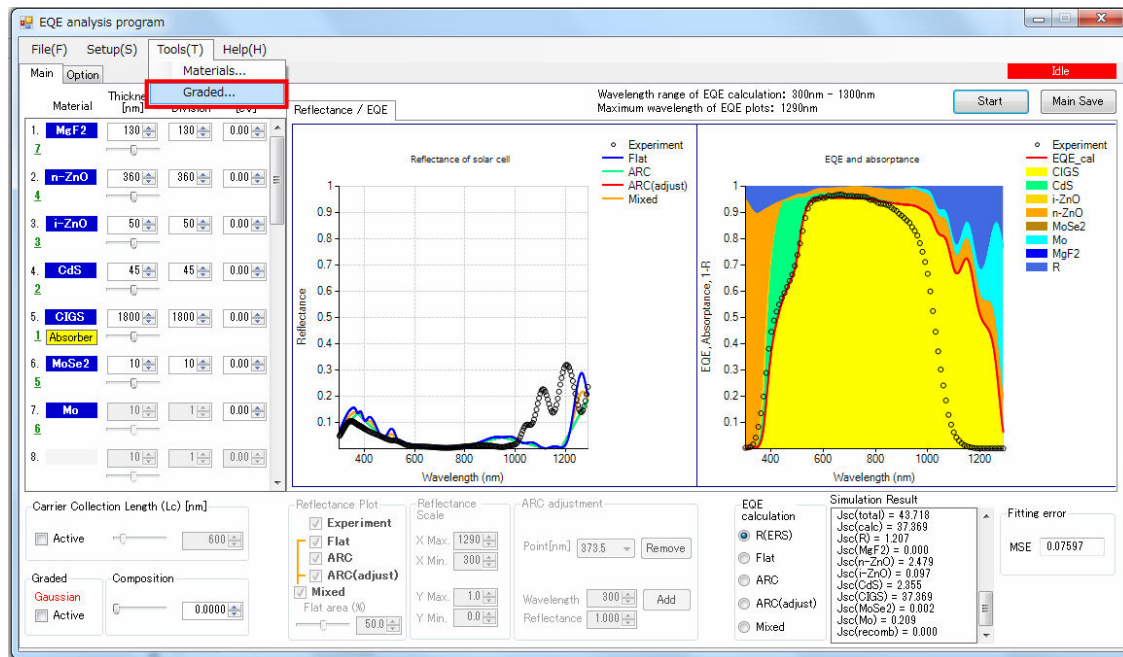


Figure 5.3 Parameters of the double Gaussian model [3].

### ③ Modeling of the graded structure

To set the above graded structure, click the tab menu of “Tool” and further select “Graded” menu (see below).



In the generated window, first select the model (“Linear” or “Gaussian”). In the linear model, input first a total number of model data points (Insert the value to the “Points” text box). Then set the composition  $x$  at each  $d$ . In this case, the  $d$  values need to be set incrementally. The calculation result can be confirmed in the plot of the “Graded layer” window. In the Gaussian mode, after the six parameter values are set, the compositional profile is also shown automatically. If the parameters are fine, click the “OK” button. The profile text data can also be saved and be loaded using “Save” and “Load” buttons.



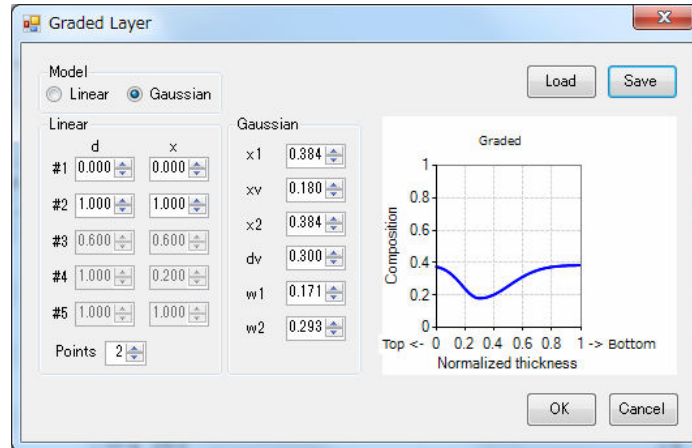


Figure 5.4 Graded layer screen

#### ④ Analysis result of the CIGSe solar cell

Figure 5.5 shows the analysis result obtained for the CIGSe solar cell reported in Ref. [3]. In this analysis, the Gaussian model with the parameters shown in Fig. 5.4 is used. It can be seen that the calculated spectrum shows excellent agreement with the experimental result with a very low MSE value.

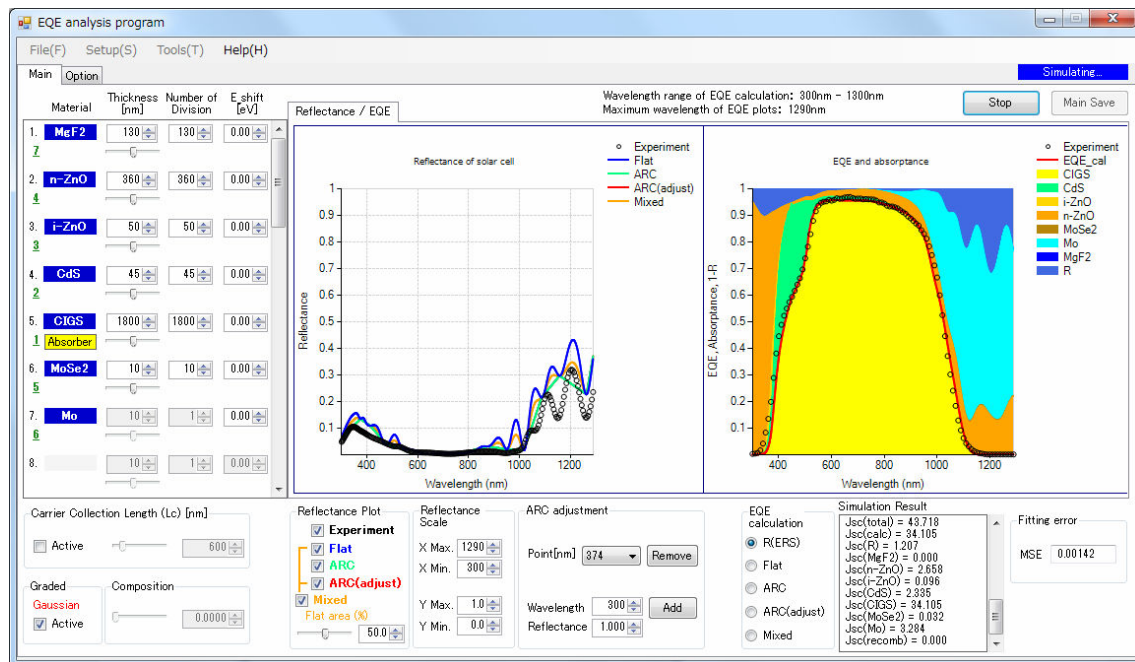


Figure 5.5 Analysis result obtained for the CIGSe solar cell reported in Ref. [3].



## 6. Example 3: EQE simulation of a superstrate-type hybrid perovskite solar cell

In this section, the EQE spectrum of a standard superstrate structure of a  $\text{CH}_3\text{NH}_3\text{PbI}_3$  (MAPbI<sub>3</sub>) solar cell fabricated with a thick glass substrate (a few mm) is simulated. In a superstrate-type solar cell with a thick glass substrate, incoherent superposition of light wave occurs and the EQE calculation needs to be performed under the incoherent condition [1]. In the e-ARC software, the incoherent optical transmission can be reproduced explicitly using a procedure of Ref. [1].

### 6.1 Setting of optical model

Figure 6.1 shows the window of “Layer Model Configuration” (see Section 4.2) with a setting for a MAPbI<sub>3</sub> solar cell. The detailed EQE analysis of MAPbI<sub>3</sub> solar cells is described in Ref. [2] and a MAPbI<sub>3</sub> solar cell structure of Glass/SnO<sub>2</sub>:F/TiO<sub>2</sub>/MAPbI<sub>3</sub>/sprio-OMeTAD /Ag is assumed.

The optical response of the optically thick transparent substrate (incoherent layer) can be set within the “Layer Model Configuration” window. Click the check box of the “Glass substrate” subwindow if the optically thick (typically >100  $\mu\text{m}$ ) incoherent layer (or substrate) is incorporated into solar cells, as shown below. Although the name of the subwindow is “Glass sub.”, other transparent materials can also be selected as an incoherent layer by clicking the button.

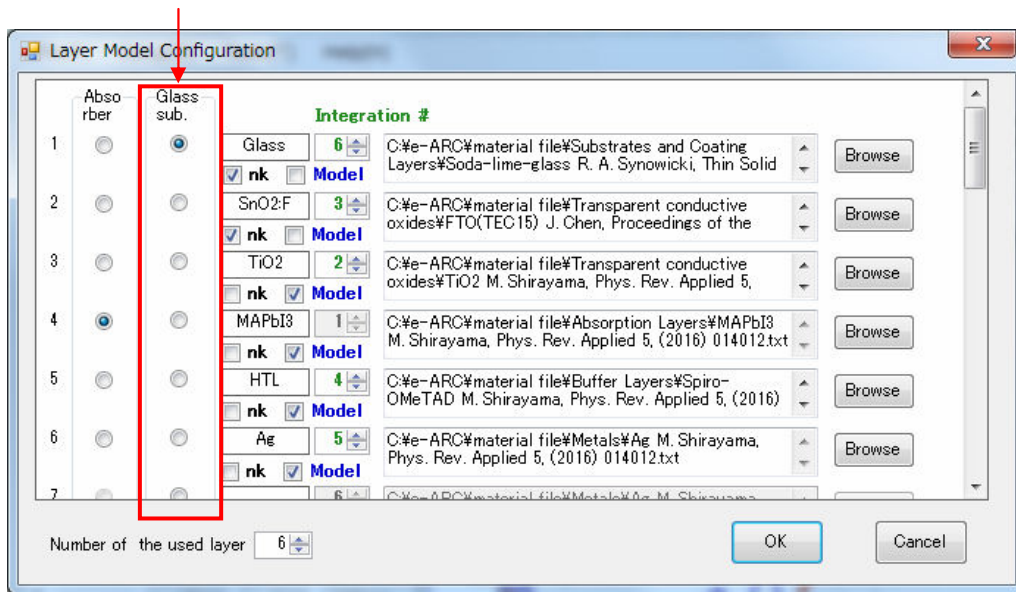


Figure 6.1 Screen of “Layer Model Configuration” window prepared for the analysis of MAPbI<sub>3</sub> solar cell.

## 6.2 Simulation result of the MAPbI<sub>3</sub> solar cell

Figure 6.2 shows the screen obtained after the EQE calculation described in Ref. [2]. In window A, the yellow “Glass” indicator is shown for the glass substrate (see the red square in the figure). This indicator shows that the optical response in this layer is calculated assuming incoherent condition.

In the model of Fig. 6.2, a dead layer (4 nm) is provided at the TiO<sub>2</sub>/MAPbI<sub>3</sub> interface due to the intense carrier recombination in this region [2]. By incorporating this dead layer, the agreement with the experimental results improves significantly.

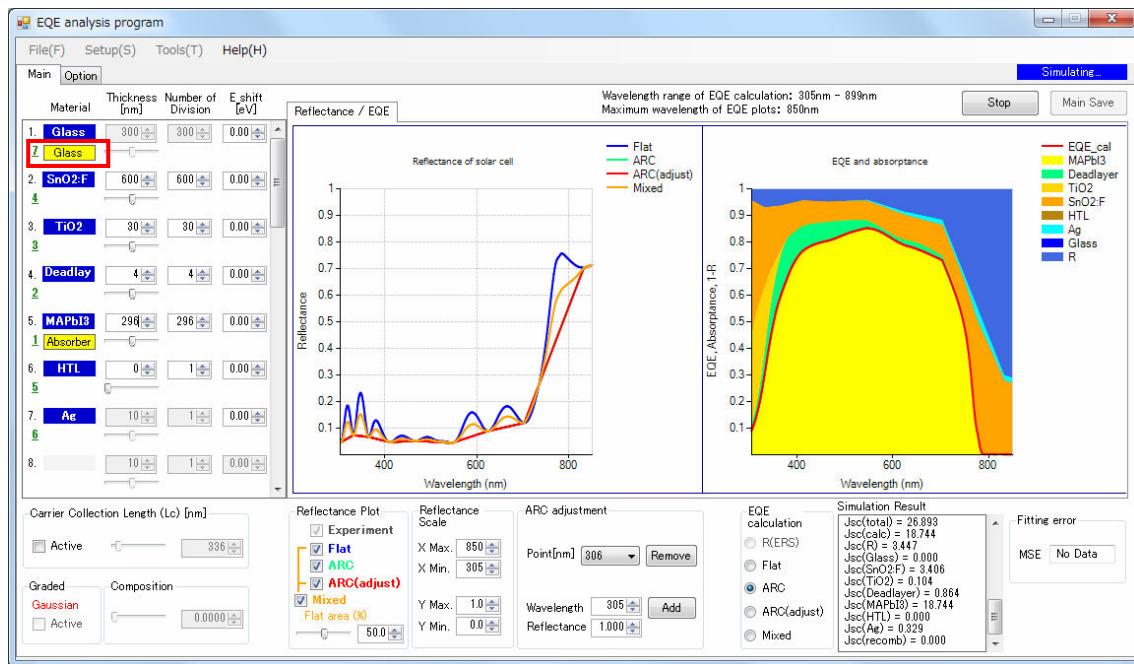


Figure 6.2 Screen obtained after the EQE analysis described in Ref. [2].

## 7. Calculation of (n, k) data from dielectric-function model parameters

Using a special feature of this software, (n, k) data can be calculated from dielectric-function model parameters. In particular, the parameters of the Tauc-Lorentz, Drude, and Sellmeier models have been summarized for numerous solar-cell component layers (see Ref. [1]) and, by applying these parameters, numerical (n, k) spectra can be obtained.

This can be implemented by clicking the “Tool” tab menu, followed by the selection of “Material” tab menu. By this procedure, “Create nk data” window is generated. If “Load Model Parameters” button is clicked, a “.txt” file can be selected and the parameter values of the dielectric function model are loaded automatically. In this case, the calculation is performed to obtain the corresponding (n, k) spectra. The parameter values found in the literature can be inputted directly by typing the numerical values in the window. When this is implemented, the number of the peaks (models) also needs to be selected at the bottom of the window. The units of the model parameters can be found in Table 4.1 (p. 15). After clicking the “Calculate/Save” button, the (n, k) output spectra are generated. Note that, when the (n, k) data created by the above procedure are used in the analysis, the energy shift feature cannot be used, as mentioned earlier.

The screenshot shows the 'Create nk data' window. It features a table with 10 rows for different peaks. Each row has a 'Peak#' column, a 'Model' dropdown menu (all set to 'TL'), and seven input fields for parameters: A/Ad/B1/A, C/G/B2/B, E0/B3/C, Eg/C1/D, e1off/C2, and C3. The first row is highlighted in blue. At the top right, there is a 'Load Model parameters' button. At the bottom, there is a 'Number of the peaks' input field (set to 10), a 'Wavelength range of calculation' input field (set to 280 [nm] ~ 3999 [nm]), and a 'Calculate / Save' button. The 'Load Model parameters' and 'Calculate / Save' buttons are highlighted with red boxes.

Peak#	Model	A/Ad/B1/A	C/G/B2/B	E0/B3/C	Eg/C1/D	e1off/C2	C3
1	TL	0	0	0	0	0	0
2	TL	0	0	0	0	0	0
3	TL	0	0	0	0	0	0
4	TL	0	0	0	0	0	0
5	TL	0	0	0	0	0	0
6	TL	0	0	0	0	0	0
7	TL	0	0	0	0	0	0
8	TL	0	0	0	0	0	0
9	TL	0	0	0	0	0	0
10	TL	0	0	0	0	0	0

Number of the peaks: 10

Wavelength range of calculation: 280 [nm] ~ 3999 [nm]

Figure 7.1 Create nk data screen

## 8. Reference

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## 9. Biographical information of the e-ARC software

For the e-ARC software, the original software was prepared by A. Nakane (Gifu Univ.) in 2015 using a MATLAB source code. This code was translated into a C<sup>#</sup>-based software by H. Tampo (AIST) in 2017. The C<sup>#</sup> source code was further modified, to include graded structure and incoherent calculations, and perfected by S. Fujimoto (Gifu Univ.). The manual was prepared by H. Fujiwara and S. Fujimoto (Gifu Univ.). The version 1.0 was released in September 25, 2017 at EU PVSC conference. The correction made for version 1.1 was carried out by H. Tampo (AIST).

### *Version Information*

Version 1.0: Original software (Sep. 25, 2017)

Version 1.1: Software operation under different operating systems with different languages was established (May 10, 2018).