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Converting sunlight into red light in fluorosilicate glass for amorphous silicon solar cells

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ABSTRACT

Fluorosilicate glass was prepared by high-temperature melting method to explore highly efficient luminescence materials for amorphous silicon solar cells. Absorption, excitation and emission spectra of the glass were measured. The optical characters of the glass were discussed in details. The glass can efficiently convert sunlight into red light. Our glass can be applied to amorphous silicon solar cells as a converter of solar spectrum.

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1. Introduction

To solve energy and environment problems, solar energy has attracted more attention. In recent years, solar cells have been widely studied [1–6]. However, the efficiency of solar cells is still low. A major reason limiting the conversion efficiency of solar cells is their insensitivity to a full solar spectrum. For amorphous Si solar cells (Eg=1.75 eV) [7,8], the most sensitive responding light is red light. The photons with energy higher than the bandgap can be absorbed, but the excess energy will be released as heat. The photons with energy lower than the bandgap cannot be absorb. For improving the absorption efficiency of amorphous Si solar cell, it is very necessary to turn sunlight into red light. Red luminescent materials doped with rare-earth and transition metal ions have been widely studied [9–16]. However, it is unusual that the materials can turn violet, visible, and infrared photons into red photons. Fluorosilicate glass has lower phonon energy ($\sim$926 cm^{-1}) compared with that of silicate glass ($\sim$1200 cm^{-1}), which is helpful to decrease the multiphonon relaxations and improve the luminescent efficiency of luminescent ions. And the fluorosilicate glass has good thermal stability. The thermal parameters of the glass (Tg, Tx…) can be referred in reference [17]. Eu^{3+} ion doped fluorosilicate glasses can efficiently turn the blue, violet and UV photons into the red photons. Yb^{3+}/Er^{3+} codoped fluorosilicate glasses can emit strong up-conversion emission under infrared excitation. Mn^{2+} ion have the wide absorption bands in the blue, violet and UV, which is helpful for improving the efficiency of down-conversion emission. In this work, we prepared SiO_2-MgF_2-AlF_3-CaF_2-MnO-Eu_2O_3-Er_2O_3-Yb_2O_3 glass using high-temperature melting method. The up- and down-conversion emission spectra of the glass materials were studied in details.

2. Experimental

The fluorosilicate glass with the compositions of 50SiO_2-20MgF_2-13AlF_3-10CaF_2-2MnO-1.5Eu_2O_3-Er_2O_3-2.5Yb_2O_3 (mol%) was prepared by high-temperature melting method. The start raw materials including SiO_2, MgF_2, AlF_3, CaF_2, $MnCO_3$, Eu_2O_3, Er_2O_3, and Yb_2O_3 were mixed thoroughly and melted at 1450 °C for 2 h in a corundum crucible. The melting liquid was poured onto a preheated stainless-steel plate in air. Finally the samples were heated at 450 °C for 6 h to release the thermal stress. The excitation and down-conversion emission spectra were measured with a model F111AI fluorescence spectrophotometer under the excitation of a xenon lamp (model Xe900). The up-conversion emission was measured under the excitation of 975 nm laser diode. The visible light was detected by a photomultiplier tube detector. The absorption spectra were recorded using a Shimadzu UV-2200 spectrophotometer. All measurements were taken at room temperature.

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3. Results and discussion

Fig. 1a) is the solar radiation spectrum in the 240–1200 nm wavelength region. In the solar spectrum, the energy percentages of the ultraviolet, visible, and infrared light are 7%, 50%, and 43%, respectively. Fig. 1b) is excitation and absorption spectra of the multifarious ions doped fluorosilicate glass. A wide excitation band from 250 to 550 nm monitored at 614 nm is observed. The wide excitation bands at 240–330 nm, 330–385 nm, 385–450, and 450–550 nm should come from the transitions of Mn^{2+} ions: $^6A_1g(S)$-$^4A_2g(F)$, $^6A_1g(S)$-$^4T_2g(D)$, $^6A_1g(S)$-$^4A_1g(F)$,$^4Eg(G)$, and $^6A_1g(S)$-$^4T_1g(G)$,$^4T_2g(G)$. The sharp peaks at 318, 361, 384, 393, 412, 472, and 525 nm should be from the transitions of Eu^{3+} ion:$^7F_0$-$^3P_0$, $^7F_0$-$^5D_4$, $^7F_1$-$^5L_7$, $^7F_0$-$^5L_6$, $^7F_1$-$^5D_3$,$^7F_0$-$^5D_2$, and $^7F_{0,1}$-$^5D_1$, respectively. The conclusions can be referred in the excitation spectra of single Mn^{2+}-doped and single Eu^{3+}-doped samples, seen in Fig. 2. The wide excitation bands of Mn^{2+} ion in the green, blue, violet and UV wavelength region can be observed in Fig. 2, which shows Mn^{2+} ion can efficiently absorb the solar and improve the efficiency of down-conversion emission. The absorption peak from 850 to 1040 nm should come from the transition of Yb^{3+} ion: $^2F_{7/2}\rightarrow{}^2F_{5/2}$.

Fig. 3 is down-conversion emission spectra of multifarious ions doped sample in red light under 267, 318, 361, 384, 393, 412, 472, and 525 nm excitation. The sharp peaks at 590, 614, 648, and 702 nm should come from the transitions of Eu^{3+} ions: $^5D_4\rightarrow{}^7F_{1,2,3,4}$. The wide emission band form 560 to 720 nm should be from the transitions of Mn^{2+} ion: $^4T_{1g}\rightarrow{}^6A_{1g}$. The conclusions can be referred in the emission spectra of single Mn^{2+}-doped and single Eu^{3+}-doped samples, seen in Fig. 4. The result show that the materials can efficiently turn the long wavelength photons into the short wavelength red photon, which are the efficient down-conversion red materials.

Fig. 5 is up-conversion emission spectra of the multifarious ions doped sample in the 400–750 nm region under 975 nm excitation. Strong red up-conversion emissions can be observed, the corresponding transitions have been indicated in the figure. Weak green up-conversion emission at 520–560 nm are found. The power spectra of the up-conversion emissions were measured under 975 nm excitation.The log-log plot for the dependence of the emission intensities on pump power is shown in the inset of Fig. 5. According to the formula [18]: $I_{up} = P^n$, where I_{up} is the UC emission intensity, P is the pump laser power, and n represents the number of laser photons absorbed when emitting an UC photon.

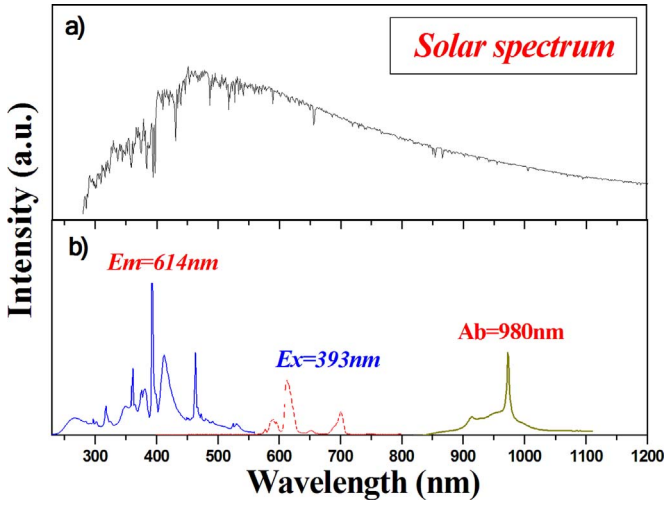

Fig. 1. a) Solar radiation spectrum in the 240–1200 nm wavelength region; b) Excitation and absorption spectra of the multifarious ions doped fluorosilicate glass.

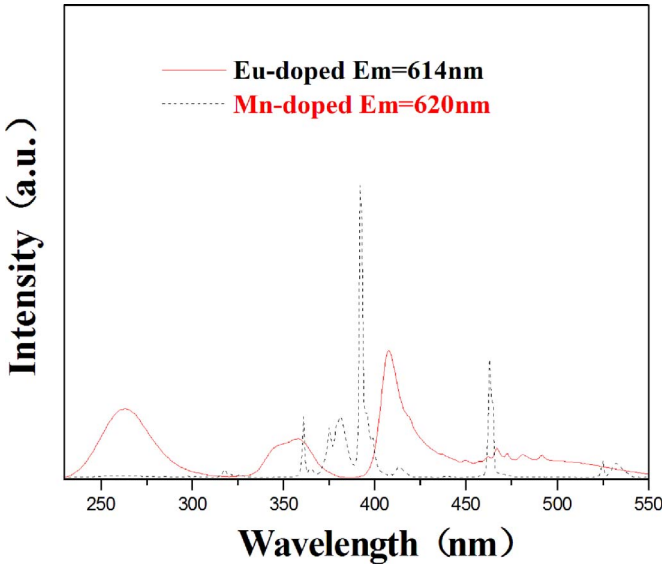

Fig. 2. Excitation spectra of single Mn^{2+}-doped and single Eu^{3+}-doped fluorosilicate glasses.

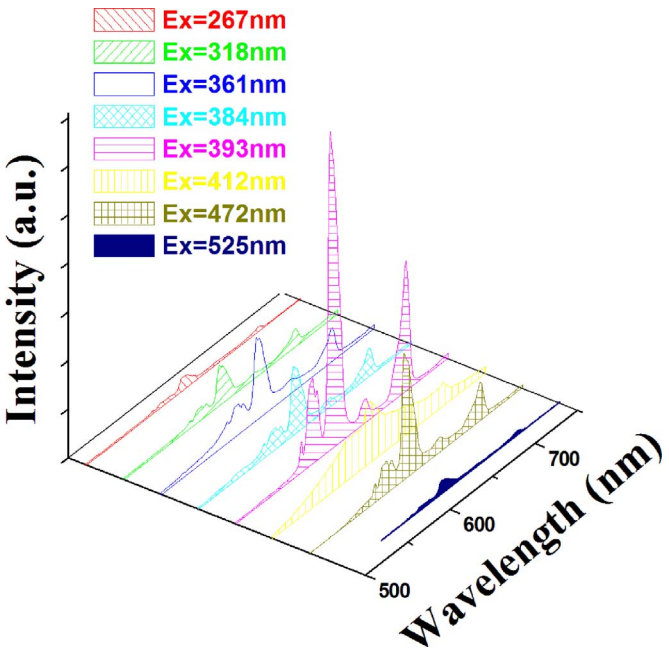

Fig. 3. Emission spectra of the multifarious ions doped fluorosilicate glass under 267, 318, 361, 384, 393, 412, 472, and 525 nm excitation.

The n values of the red, orange, and green up-conversion emissions are 1.7, 1.8, and 2.0, which indicates the two-photon processes.

Energy level diagram of Eu^{3+}, Er^{3+} and Yb^{3+}ions, as well as the proposed UC mechanism is shown in Fig. 6. The population processes of the red up-conversion emission can be described as follows. Under 975 nm excitation, the Yb^{3+} ions in the ground state $^2F_{7/2}$ were excited to the excited state $^2F_{5/2}$. By energy transition(ET) 1: $^2F_{5/2}(Yb^{3+})+{}^4I_{15/2}(Er^{3+})\rightarrow{}^2F_{7/2}(Yb^{3+})+{}^4I_{11/2}(Er^{3+})$, the Er^{3+} ions in the ground state $^4I_{15/2}$ were pumped to the excited state $^4I_{11/2}$. The Er^{3+} ions in $^4I_{11/2}$ can be excited to $^4F_{7/2}$ state by ET2: $^2F_{5/2}(Yb^{3+})+{}^4I_{11/2}(Er^{3+})\rightarrow{}^2F_{7/2}(Yb^{3+})+{}^4F_{7/2}(Er^{3+})$. At the same time, the Er^{3+} ions in $^4I_{11/2}$ can also relaxed to $^4I_{13/2}$ state by non-radiative transition. The Er^{3+} ions in $^4F_{7/2}$ state can relaxed to $^4F_{9/2}$ state by continuous multi-phonon relaxation. The Er^{3+} ions in $^4I_{13/2}$ state can be pumped to $^4F_{9/2}$ state by ET3:$^2F_{5/2}(Yb^{3+})+$

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