Effect of solvent on the deposition behavior of MOCVD-Pb(Zr,Ti)O_3 films using liquid-deliver source supply system

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Introduction

Liquid delivery source supply system, in which source materials are dissolved into the solvent and vaporized at vaporizer, has been widely used for the MOCVD preparation of Pb(Zr,Ti)O_3 films to increase the deposition rate of the films. However, the effect of the solvent has been hardly investigated. The decrease of the deposition rate and the conformability by the addition of C_4H_8O solvent were reported for Ru metal film deposition from $Ru(C_5H_4C_2H_5)_2$-O_2 system(1).

In the present study, the effect of the solvent was investigated from the viewpoint of the deposition rate and the film composition.

Experimental

Pb(Zr,Ti)O_3 film was deposited at 600 ^oC on (111)Pt/SiO_2/Si substrates from $Pb(C_{11}H_{19}O_2)_2$-$Zr(O\cdot t\text{-}C_4H_9)_4$-$Ti(O\cdot i\text{-}C_3H_7)_4$-$O_2$ system(2). These metal sources were delivered by two supply methods; conventional bubbling method and the liquid-delivery supply methods in which sources were dissolved into C_8H_8 and vaporized at individual vaporizers. The theoretical source gas flow rate was defined as *R(source)*(3) in bubbling delivery system. In the present study, $R[Pb(Pb(C_{11}H_{19}O_2)_2]$ was changed under the constant of $R[Zr(O\cdot t\text{-}C_4H_9)_4]$ and $R[Ti(O\cdot i\text{-}C_3H_7)_4]$.

The deposition rate of the constituent elements and the composition of the films were estimated by X-ray fluorescence calibrated by using the standard samples.

Results and Discussions

Fig.1 shows the change of the film composition as a function of the input gas flow ratio of Pb source to the summation of those of Zr and Ti ones. In case of bubbling delivery methods, there is the region where the stoichiometric PZT composition, Pb/(Pb+Zr+Ti)=0.5, was obtained against the $R[Pb(C_{11}H_{19}O_2)_2]$ /{ $R[Zr(O\cdot t\text{-}C_4H_9)_4]$ + $R[Ti(O\cdot i\text{-}C_3H_7)_4]$}. However, this region was not observed for the liquid delivery methods. This suggests that the strict control of the input source gas composition was essential in this case. Moreover, the deposition rate of each constituent element was ascertained to decrease even though above 10 times larger source gas was introduced into the reactor in case of the liquid delivery supply methods. This suggests that the solvent play an important role for the deposition.

To understand the effect of the solvent, the solvent vapor was added into the bubbling source system. Fig.2 shows the deposition rates of each constituent element in the films as a function of the gas flow rate of the solvent. The decrease of the deposition rate of all constituent elements was ascertained. The data by adding C_4H_8O are also shown in Fig.2, suggesting that the decrease of the deposition rate by the addition of the solvent vapor is also observed. These results clearly shows that the effect of the solvent into the deposition mechanism must be take into account for PZT film deposition from the liquid-delivery system.

References

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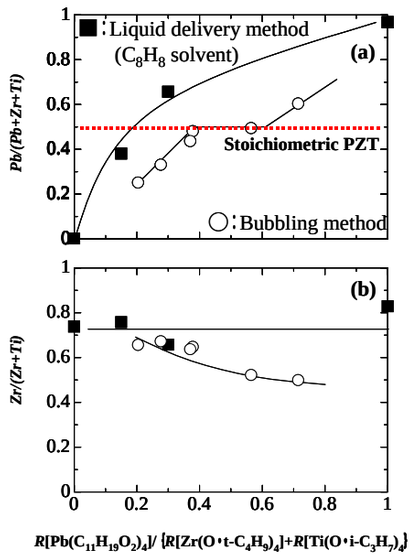

Fig.1 PZT film composition as a function of the input gas flow ratio of Pb source to the summation of those of Zr and Ti ones.

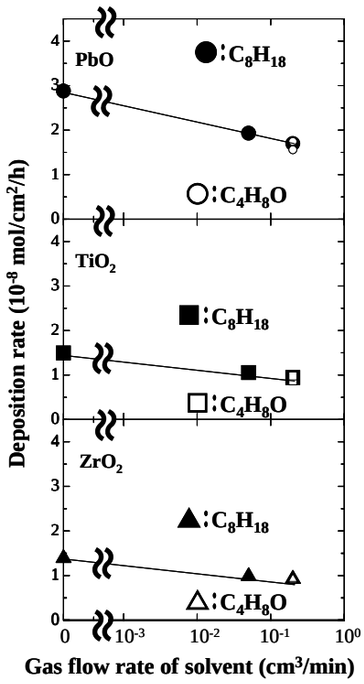

Fig.2 Deposition rates of each constituent element in PZT films as a function of the gas flow rate of solvent.