Electronic Supplementary Information for

Rational engineering of a switching material for the ovonic threshold switching (OTS) device with mitigated electroforming

Jaesang Lee[1,2], Seong Won Cho[1,2], Young Woong Lee[1,3], Joon Young Kwak[1], Jaewook Kim[1], Yeonjoo Jeong[1], Gyu Weon Hwang[1], Seongsik Park[1], SangBum Kim[2], Suyoun Lee[1,4*]

[1]*Center for Neuromorphic Engineering, Korea Institute of Science and Technology, Seoul, 02792, South Korea*

[2]*Department of Materials Science and Engineering, Seoul National University, Seoul, 08826, South Korea*

[3]*Department of Physics, Konkuk University, Seoul 05029, South Korea*

[4]*Division of Nano & Information Technology, Korea University of Science and Technology, Daejeon 34316, Korea*

* Corresponding author: slee_eels@kist.re.kr

S1. Absorption spectroscopy of GSS films

Fig. S1 shows the results of the absorption spectroscopy of GSS films in the form of the Tauc plot ($(\alpha h\nu)^n$ vs. $h\nu$, where α, h, and ν are the absorption coefficient, Planck constant, and the frequency of the incident photon, respectively. n is set to 1/2 under an assumption of the indirect semiconductors). The measurements have been performed by using a UV-Vis-NIR spectrometer (Cary 5000, Agilent.). The energy gap is obtained from the fitting line to the data in the high energy regime as can be found in Fig. S1(a). In Fig. S1(f), the obtained energy gap ($E_{g,opt}$) is plotted together with the energy gap ($E_{g,pristine}=2\times(E_C-E_F)$) obtained by using the method of the Arrhenius plot of $\Phi(T)$ shown in Fig. 2(c) in the main text.

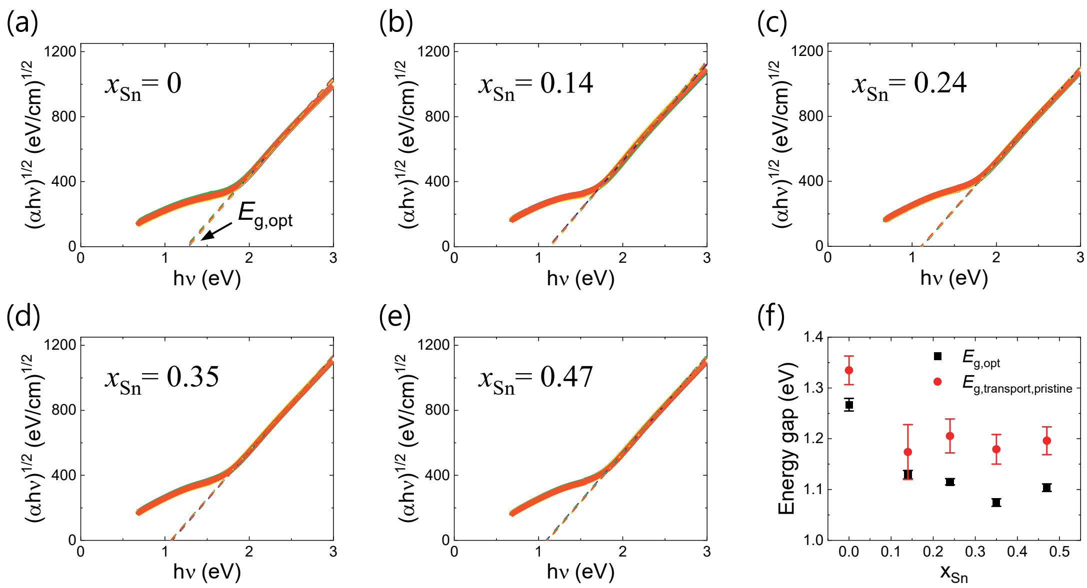

Fig. S1 (a)~(e) Absorption spectroscopy of GSS films in the form of Tauc plot ($(\alpha h\nu)^n$ vs. $h\nu$, where α, h, and ν are the absorption coefficient, Planck constant, and the frequency of the incident photon, respectively. n is set to 1/2 under an assumption of the indirect semiconductors). The measurement has been repeated five times for each sample to give very small variations (five nearly-overlapped curves in each plot). Thin dashed lines are the linear fitting curves to the experimental data (thick solid lines) in the high energy regime. (f) Energy gap ($E_{g,opt}$ and $E_{g,transport,\ pristine}$) as a function of x_{Sn}, where $E_{g,opt}$ and $E_{g,transport,\ pristine}$ are the values obtained by the absorption spectroscopy and by $\Phi(T)$ described in the main text, respectively.

S2. Switching *I-V* curve a few months after the electroforming

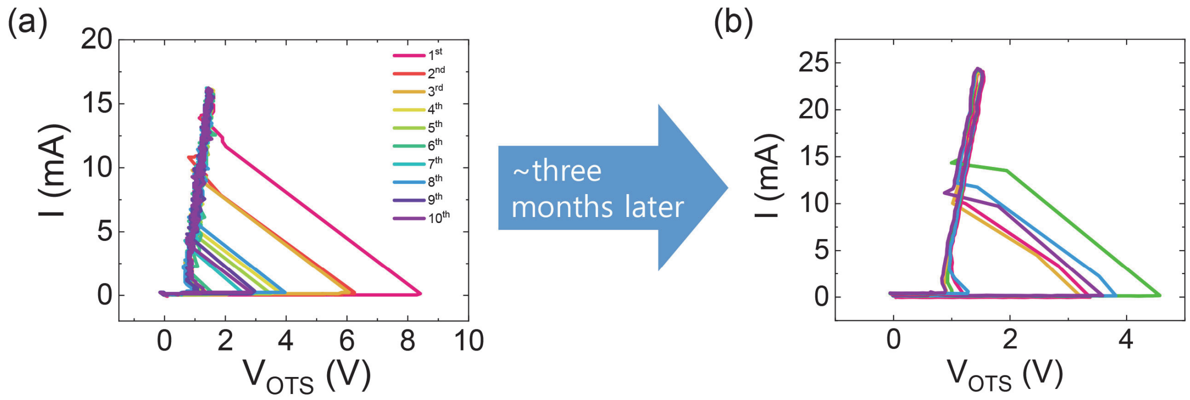

Fig. S2 Switching *I-V* curves of a representative OTS device of $Ge_{0.56}Se_{0.44}$ (a) in its pristine state (same as Fig. 1(a) in the main text) and (b) three months after the first measurement.

S3. Supplementary XPS spectra of GSS films

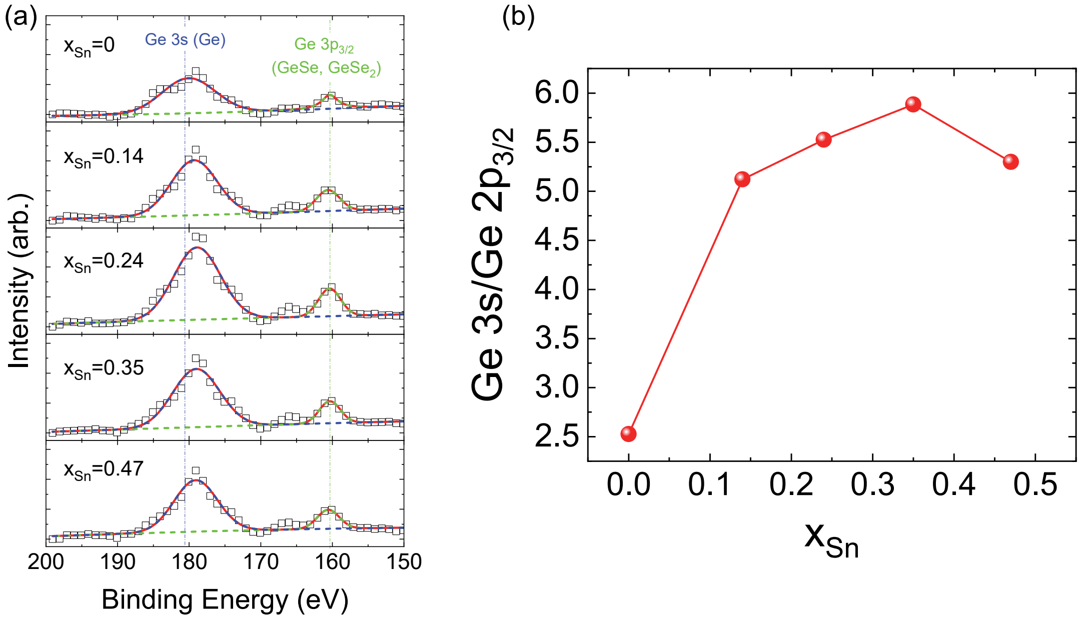

Fig. S3 (a) XPS spectra in the range of the binding energy from 150 eV to 200 eV with varying x_{Sn}. The dashed lines represent the deconvoluted peaks by the Gaussian-Lorentzian lineshape with their sum displayed as a red solid line. The vertical dash-dot lines indicate positions corresponding to Ge 3s (blue: 181 eV) and Ge $3p_{3/2}$ (green: 160.7 eV) levels, respectively. (b) The ratio of the spectral weight of Ge 3s to that of Ge 3p3/2 as a function of x_{Sn}.

S4. Switching *I-V* curves of OTS devices using GSS films

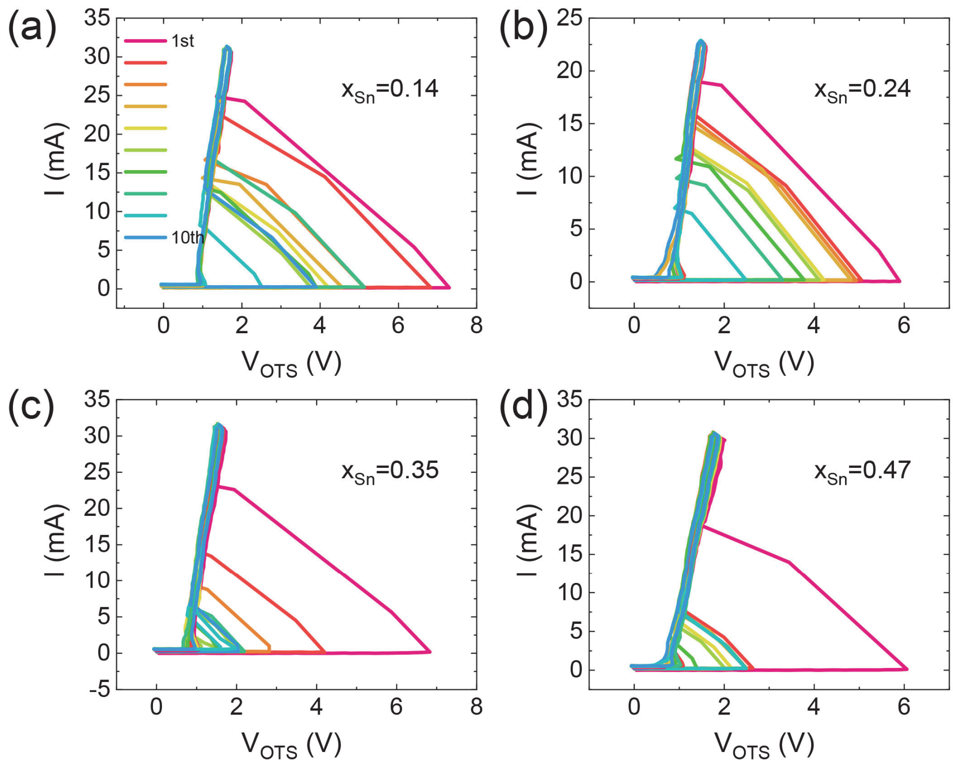

Fig. S4 (a)~(e) Evolution of the switching *I-V* curve of the OTS devices using $Sn_{0.08}Ge_{0.47}Se_{0.45}$ (a), $Sn_{0.13}Ge_{0.42}Se_{0.45}$ (b), $Sn_{0.18}Ge_{0.34}Se_{0.48}$ (c), and $Sn_{0.23}Ge_{0.26}Se_{0.51}$ (d), respectively. The color code is the same as in (a).

S5. Temperature-dependent *I-V* curves of OTS devices using GSS films in the sub-threshold regime

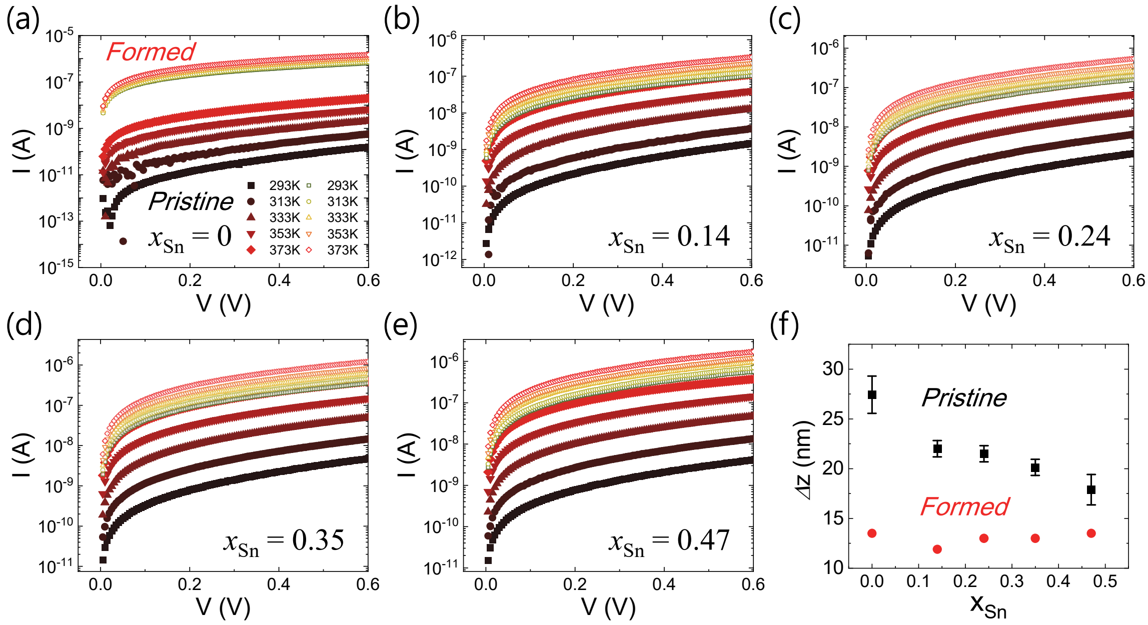

Fig. S5 (a)~(e) Temperature-dependent *I-V* curve of the OTS devices using $Ge_{0.56}Se_{0.44}$ (a), $Sn_{0.08}Ge_{0.47}Se_{0.45}$ (b), $Sn_{0.13}Ge_{0.42}Se_{0.45}$ (c), $Sn_{0.18}Ge_{0.34}Se_{0.48}$ (d), and $Sn_{0.23}Ge_{0.26}Se_{0.51}$ (e), respectively. (f) Inter-trap distance (Δz) as a function of x_{Sn}. The errorbar represents the standard deviation of five samples.

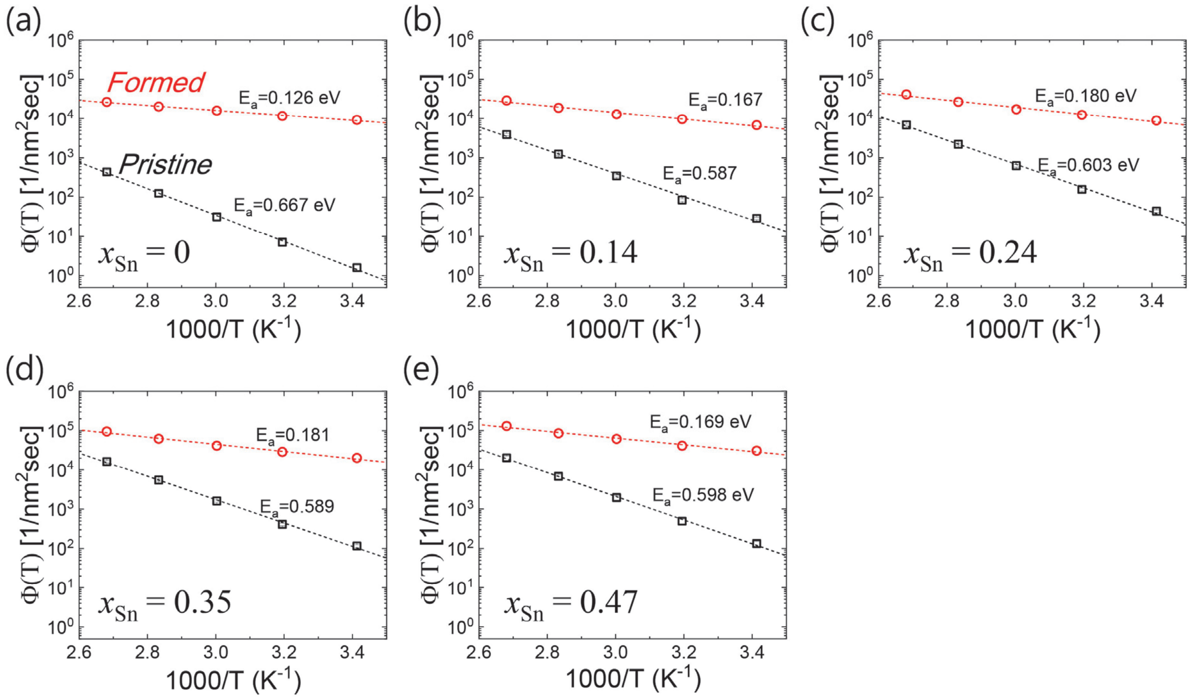

Fig. S6 (a)~(e) Arrhenius plot of $\Phi(T)$, the flux of trap states (for the definition of $\Phi(T)$, see the main text) for the OTS devices using $Ge_{0.56}Se_{0.44}$ (a), $Sn_{0.08}Ge_{0.47}Se_{0.45}$ (b), $Sn_{0.13}Ge_{0.42}Se_{0.45}$ (c), $Sn_{0.18}Ge_{0.34}Se_{0.48}$ (d), and $Sn_{0.23}Ge_{0.26}Se_{0.51}$ (e), respectively.