

Supplementary of “Structural Control of Molybdenum Oxide by post-treatment processes”

Makoto Kashiwagi, Shota Chomei, Takehiko Yokomori, Yuki Oguchi, Yuzo Shigesato

Optical property of a-MoO₃ and irradiation spectrum of flash lamp

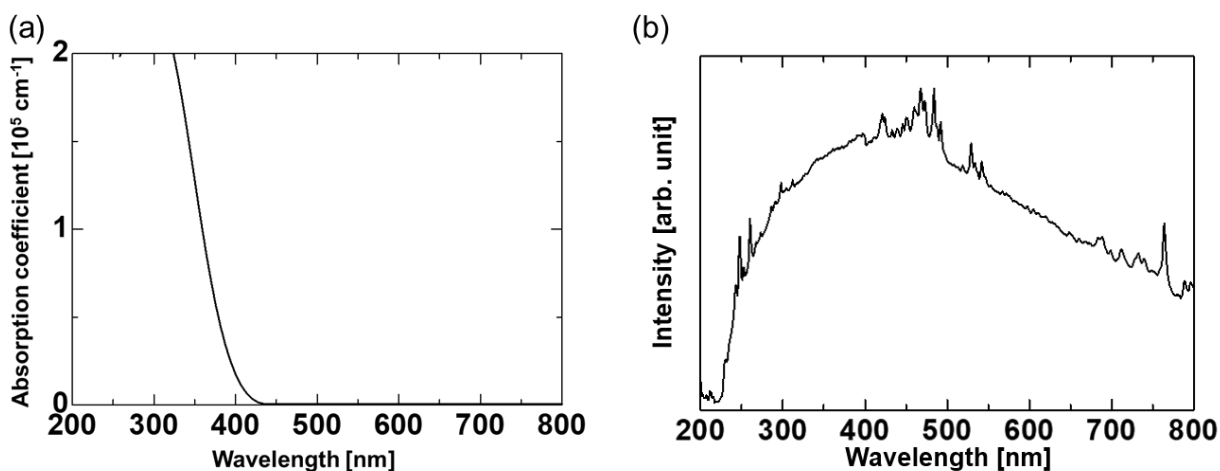


Figure S1 Optical property of a-MoO₃ thin film and irradiation spectrum of flash lamp. (a) the absorption coefficient of a-MoO₃ thin film evaluated by using spectroscopic ellipsometry. (b) the flash lamp's irradiation spectrum.

Alt text: Two graphs showing the optical properties of the film and the flash lamp: an absorption spectrum with a sharp increase below 400 nm and a broad emission spectrum of the lamp from 230 to 800 nm.

Structural analysis by X-ray diffraction and Raman Spectroscopy

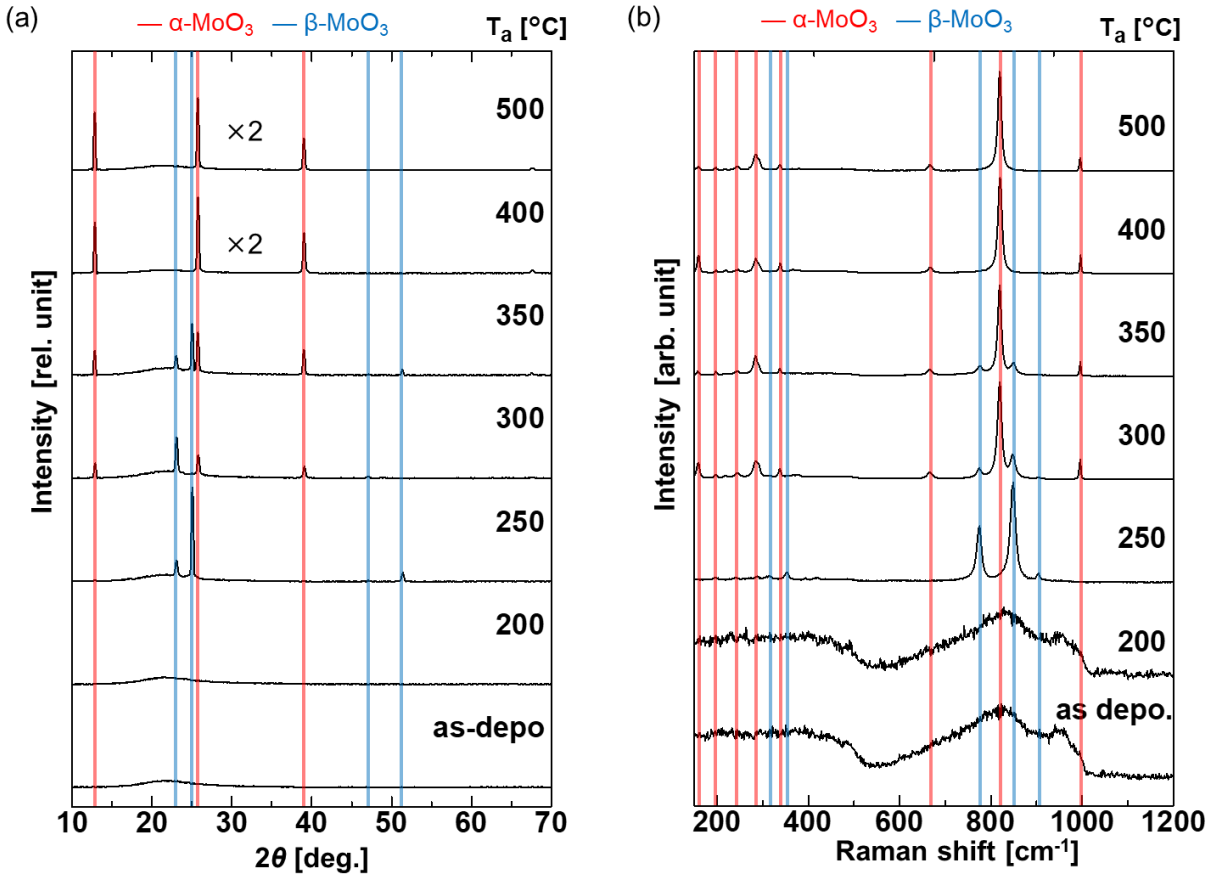


Figure S2 XRD pattern and Raman spectra of the post-thermal annealed α - MoO_3 with annealing time of 60 min. The red-lines show the peaks corresponding to α - MoO_3 and the blue-lines show the peaks corresponding to β - MoO_3 . (a) XRD pattern, (b) Raman spectra.

Alt text: Comprehensive XRD and Raman data sets for samples annealed from 200 to 500 °C, showing a wider 2θ range than Figure 1 to confirm the phase transition.

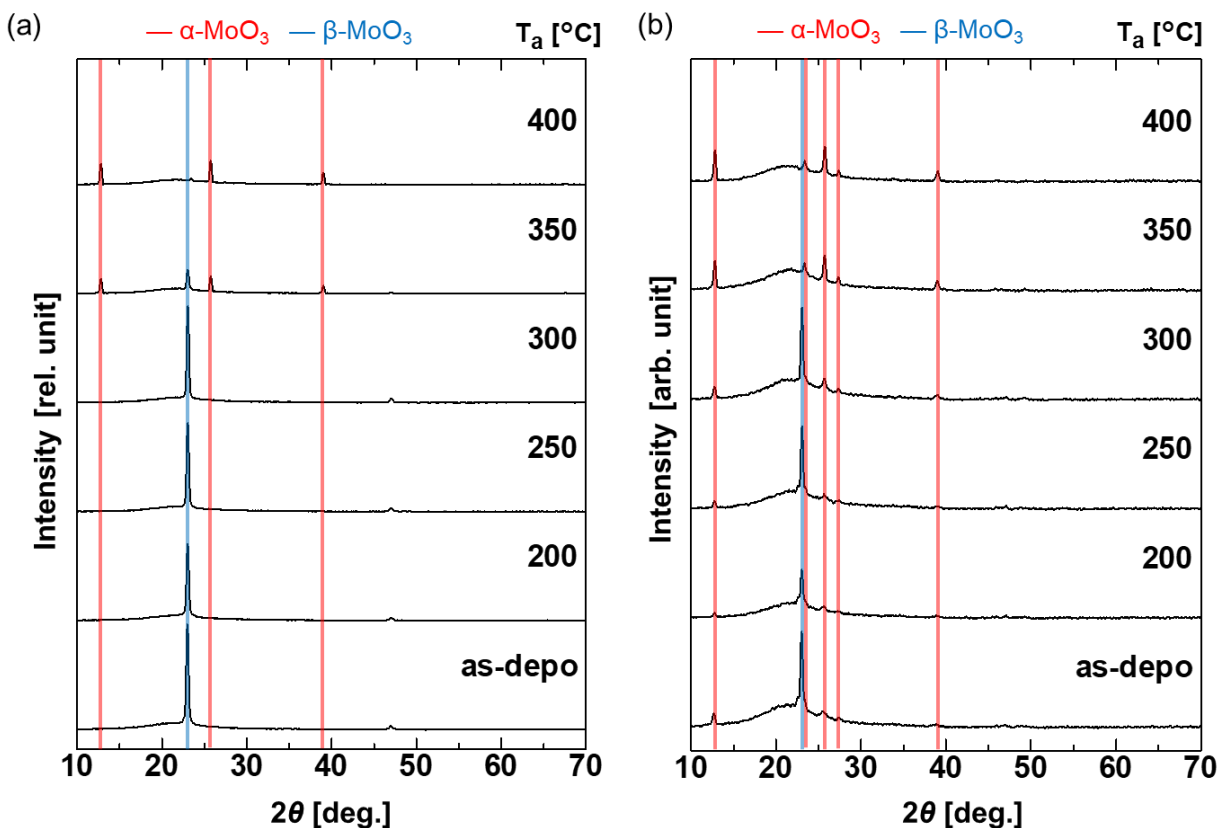


Figure S3 XRD pattern of the post-thermal annealed polycrystalline MoO_3 with annealing time of 60 min. The red-lines show the peaks corresponding to $\alpha\text{-MoO}_3$ and the blue-lines show the peaks corresponding to $\beta\text{-MoO}_3$. (a) The polycrystalline MoO_3 deposited with the substrate heating at 250 °C. (b) The polycrystalline MoO_3 deposited with the substrate heating at 350 °C.

Alt text: XRD patterns over a broad angular range for polycrystalline films deposited at 250 and 350 °C, illustrating the transition from β to α phase across various annealing temperatures from 200 °C to 400°C.

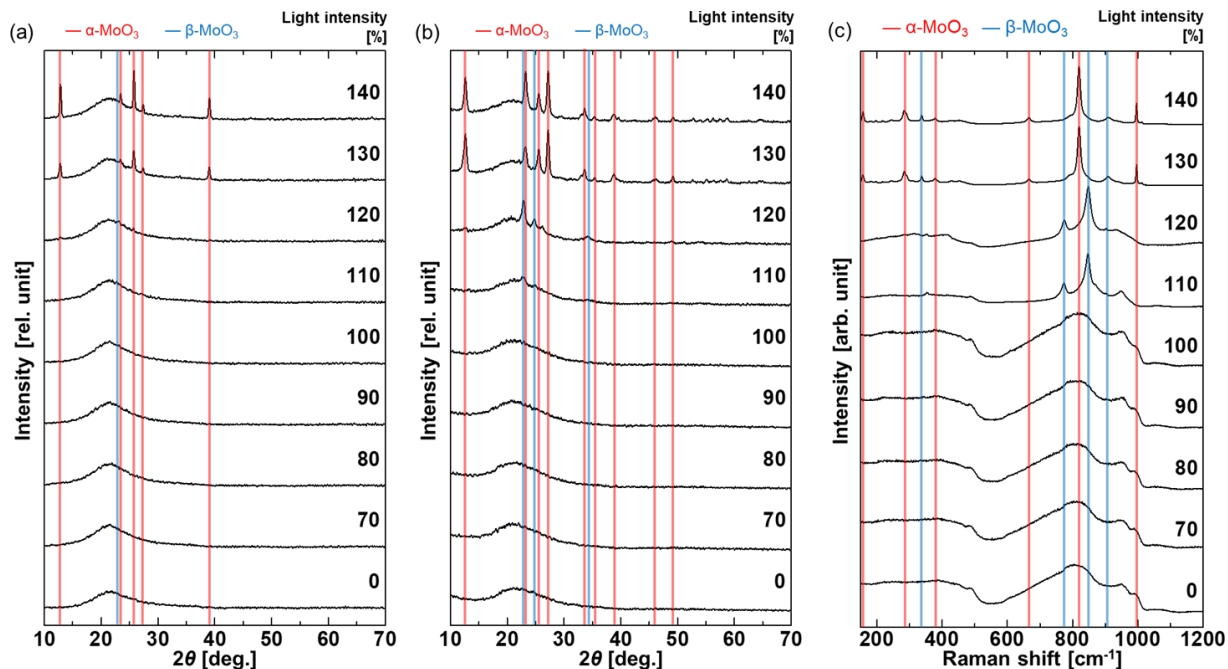


Figure S4 XRD pattern and Raman spectra of the post-flash lamp annealed a-MoO₃ with irradiation time of 0.2 ms. The red-lines show the peaks corresponding to α -MoO₃ and the blue-lines show the peaks corresponding to β -MoO₃. (a) XRD pattern (θ -2 θ scan), (b) XRD pattern (2 θ scan), (c) Raman spectra.

Alt text: Extended structural data for amorphous films after flash lamp annealing at 70% to 140% intensity, providing a complete view of the diffraction peaks for β and α phases.

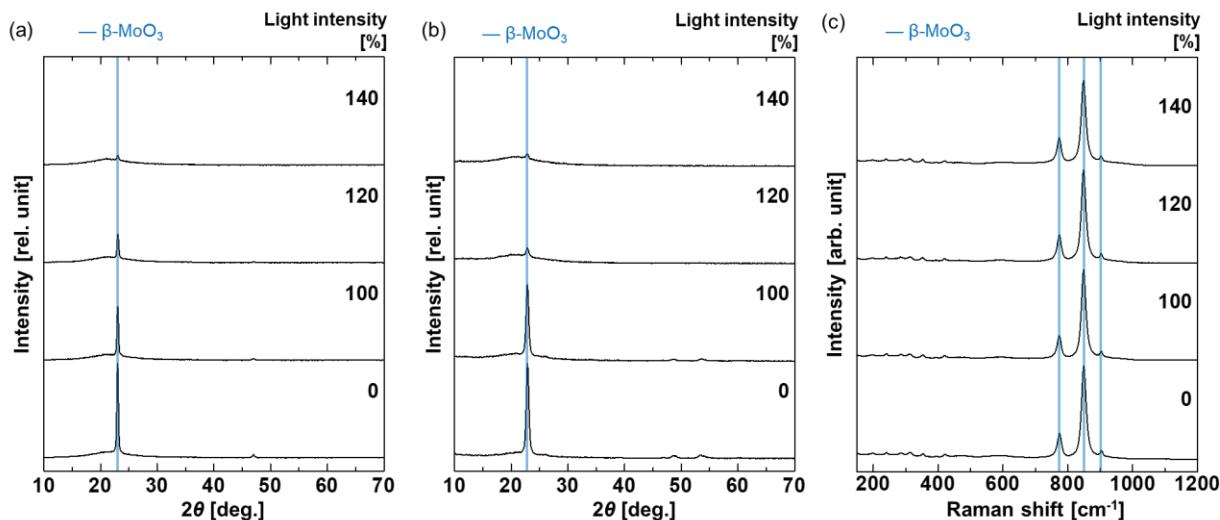


Figure S5 XRD pattern and Raman spectra of the post-flash lamp annealed polycrystalline MoO₃ with irradiation time of 0.2 ms. The polycrystalline MoO₃ thin films were deposited with the substrate heating at 250 °C. The red-lines show the peaks corresponding to α -MoO₃ and the

blue-lines show the peaks corresponding to β -MoO₃. (a) XRD pattern (θ - 2θ scan), (b) XRD pattern (2θ scan), (c) Raman spectra.

Alt text: The results of structural analysis for full measurement range of polycrystalline films after FLA show the stability of the β phase up to an intensity of 140%.

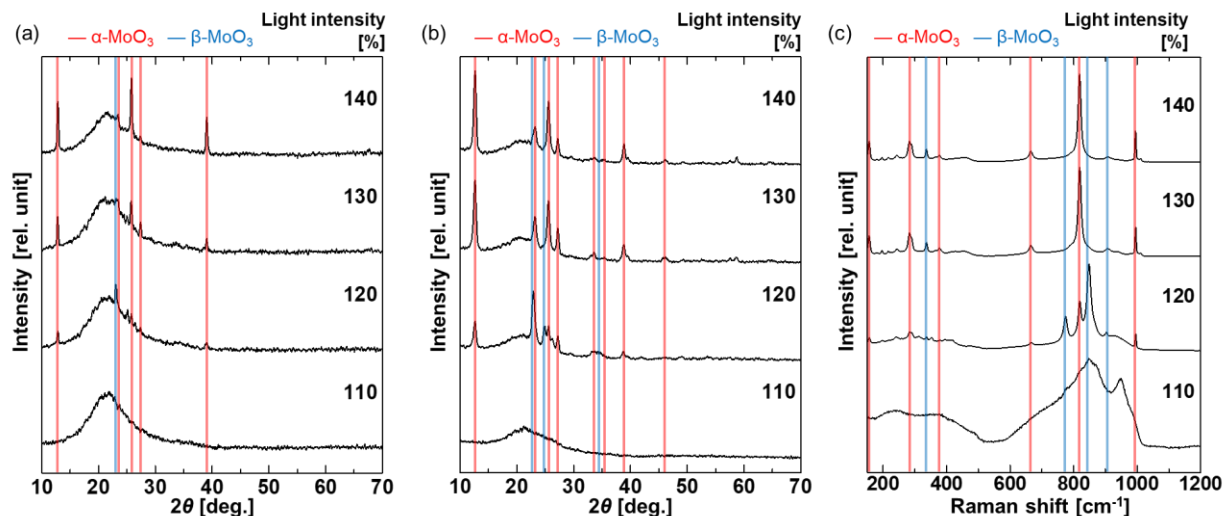


Figure S6 XRD pattern and Raman spectra of the post-flash lamp annealed a-MoO₃ with cooling by liquid nitrogen. The red-lines show the peaks corresponding to α -MoO₃ and the blue-lines show the peaks corresponding to β -MoO₃. (a) XRD pattern (θ - 2θ scan), (b) XRD pattern (2θ scan), (c) Raman spectra.

Alt text: Structural analysis results for the full measurement range of cooled samples after FLA demonstrated consistent crystallization behavior, regardless of the initial substrate temperature.

Structural analysis by Transmission Electron Microscopy

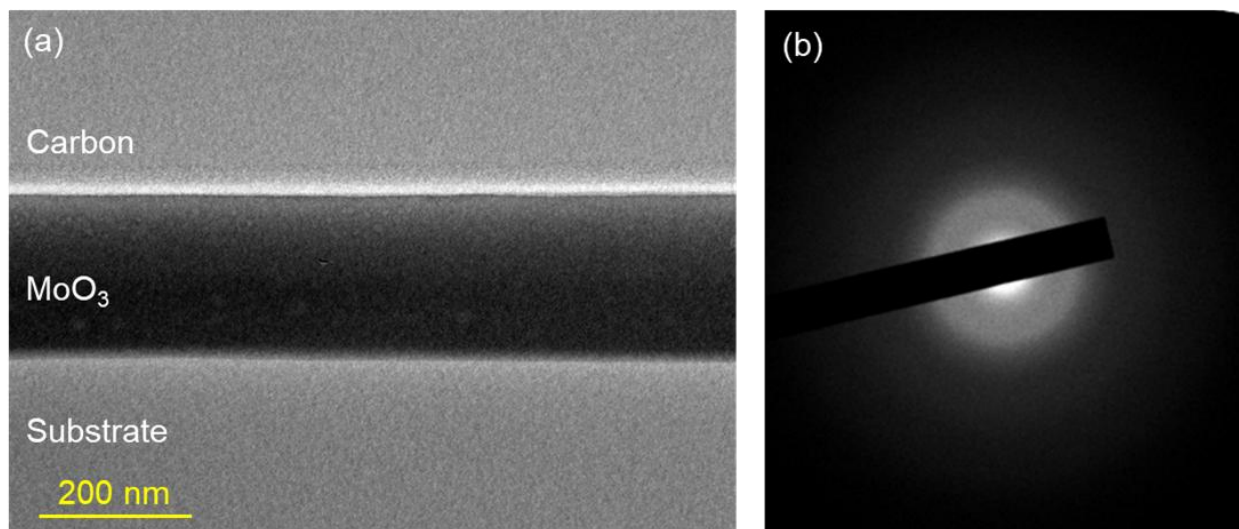


Figure S7 TEM image and electron diffraction image of as-deposited MoO₃ without substrate heating during deposition. (a) TEM image of a-MoO₃, (b) Electron diffraction image of a-MoO₃.

Alt text: The cross-sectional TEM image shows a uniform amorphous layer and an electron diffraction pattern with diffuse halos. This confirms the amorphous state of the as-deposited MoO₃.

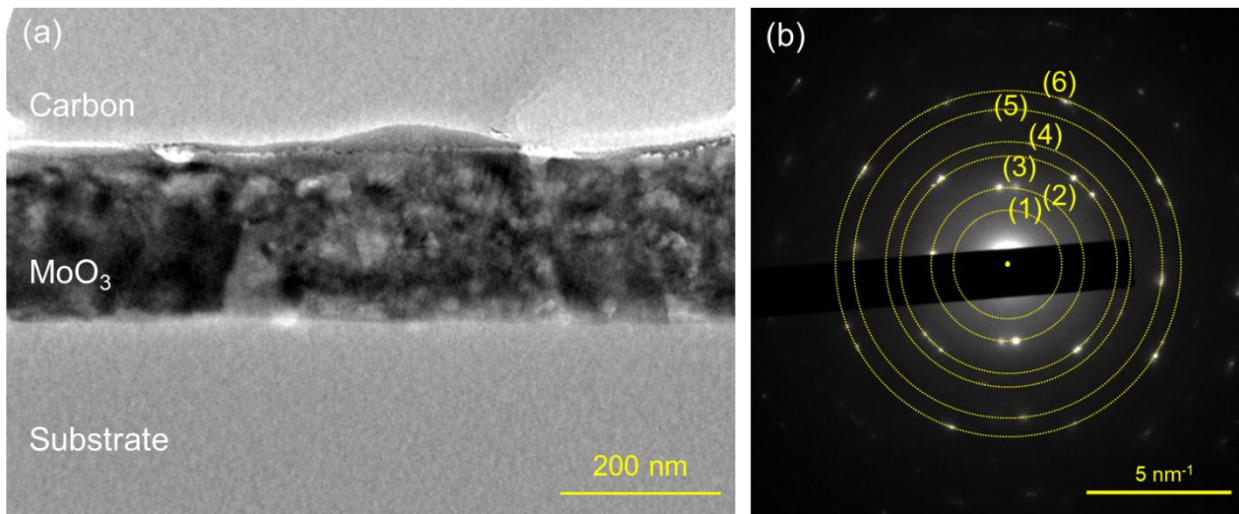


Figure S8 TEM image and electron diffraction image of post-thermal annealed a-MoO₃ with annealing time of 60 min and annealing temperature of 250 °C. (a) TEM image, (b) Electron diffraction image. The diffraction patterns (1) through (6) shown in the figure are derived from β -MoO₃(010), β -MoO₃(011), β -MoO₃(020), β -MoO₃(112), β -MoO₃(022), and β -MoO₃(031), respectively.

Alt text: The TEM image shows a crystallized layer and an electron diffraction pattern with rings that are indexed to the β - MoO_3 phase. This confirms that crystallization occurred after post-thermal annealing at 250 °C.

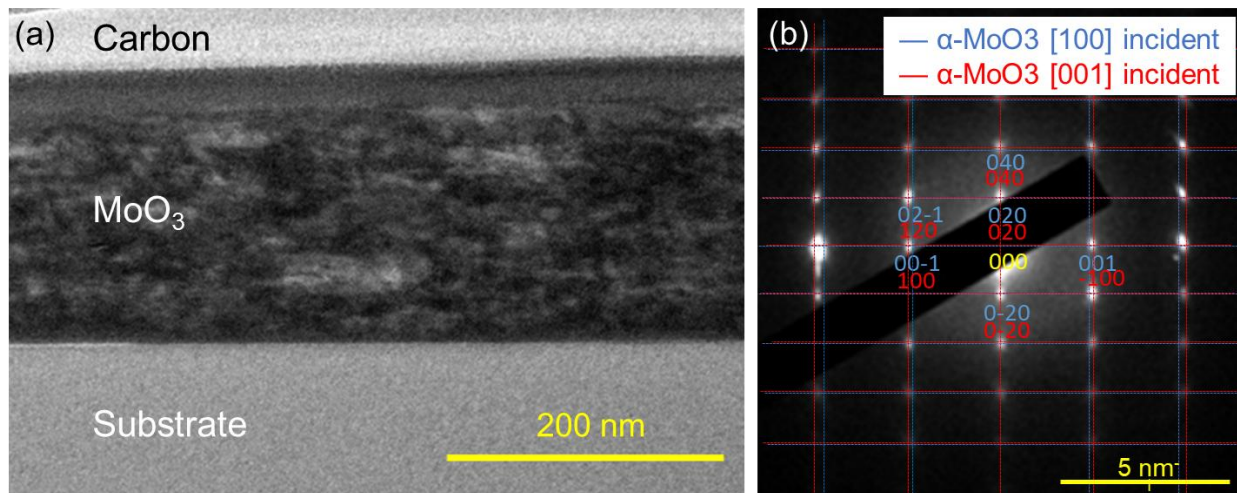


Figure S9 TEM image and electron diffraction image of post-thermal annealed α - MoO_3 with annealing time of 60 min and annealing temperature of 500 °C. (a) TEM image, (b) Electron diffraction image. The diffraction patterns in the figure are derived from α - MoO_3 .

Alt text: The cross-sectional TEM image clearly shows grains and an electron diffraction spot pattern indexed to the α - MoO_3 structure after post-thermal annealing at 500 °C.

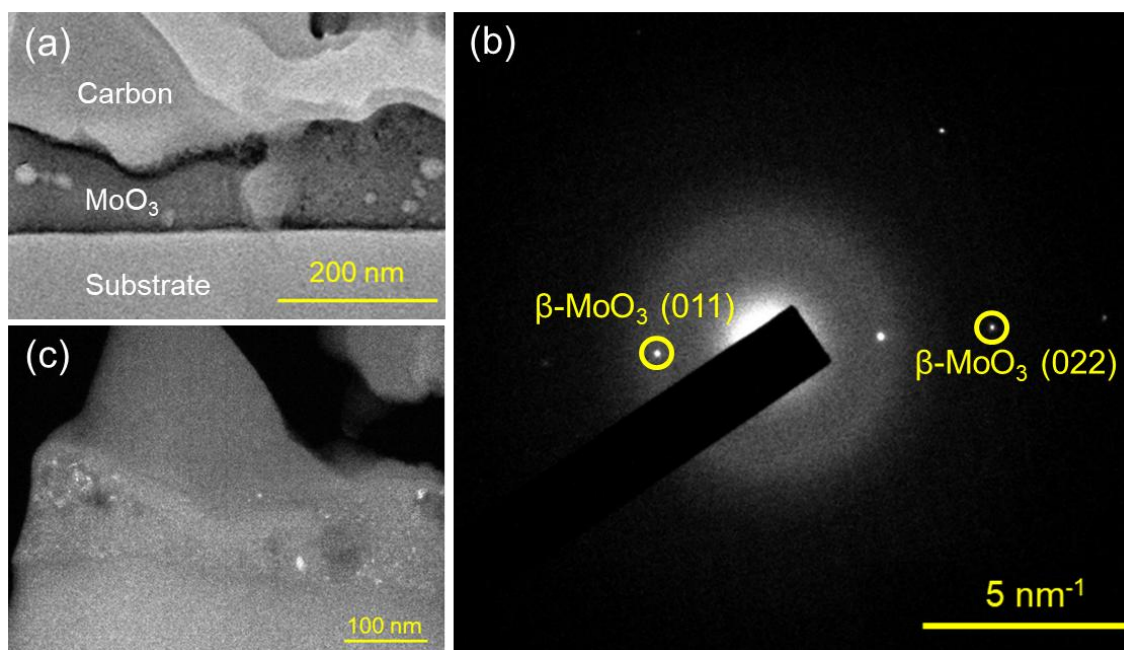


Figure S10 TEM image and electron diffraction image of post-flash lamp annealed α -MoO₃ with irradiation light intensity of 120 %. (a) bright field TEM image. (b) Electron diffraction image. (c) dark field TEM imaged regarded from β -MoO₃(011).

Alt text: The bright-field and the dark-field TEM image shows crystallization of α -MoO₃ by post-flash lamp annealing. The electron diffraction pattern is indexed to the β -MoO₃ phase.

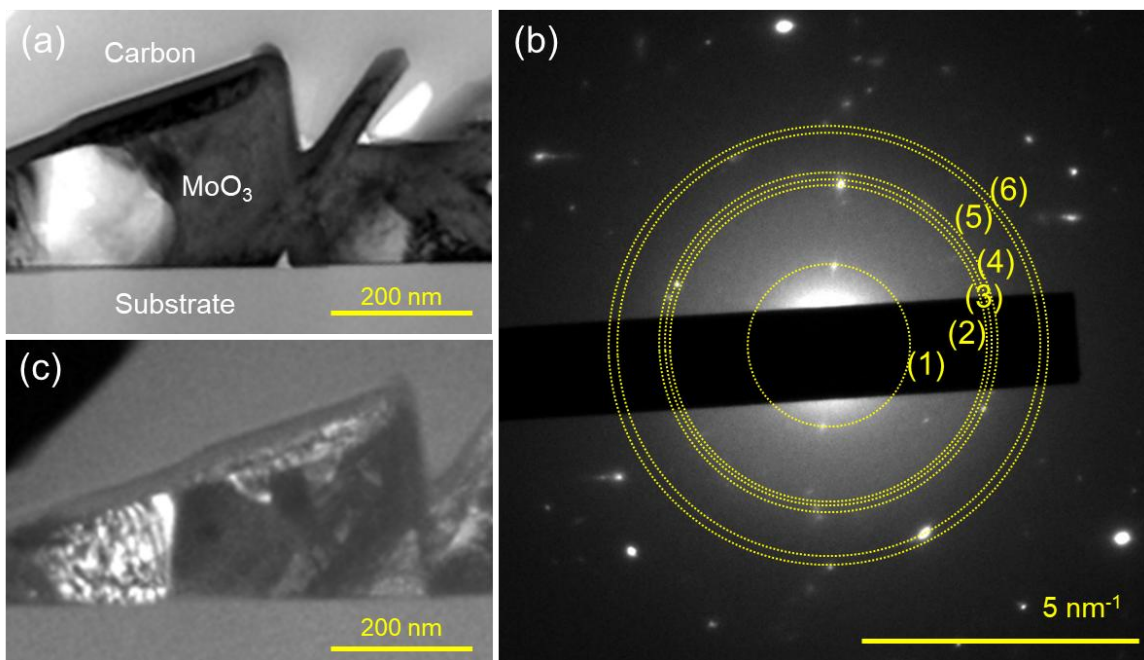


Figure S11 TEM image and electron diffraction image of post-flash lamp annealed α -MoO₃ with irradiation light intensity of 140 %. (a) bright field TEM image. (b) Electron diffraction image. The diffraction patterns (1) through (6) shown in the figure are derived from α -MoO₃(020), α -MoO₃(110), α -MoO₃(040), α -MoO₃(021), α -MoO₃(111), and α -MoO₃(041), respectively. (c) dark field TEM imaged regarded from α -MoO₃(040).

Alt text: The TEM images clearly show the crystalline structure of MoO₃. The electron diffraction ring pattern is indexed to the α -MoO₃ phase after post-flash lamp annealing.

Activation Energy Diagram of Crystallization and Phase Transition.

E_a : Activation Energy

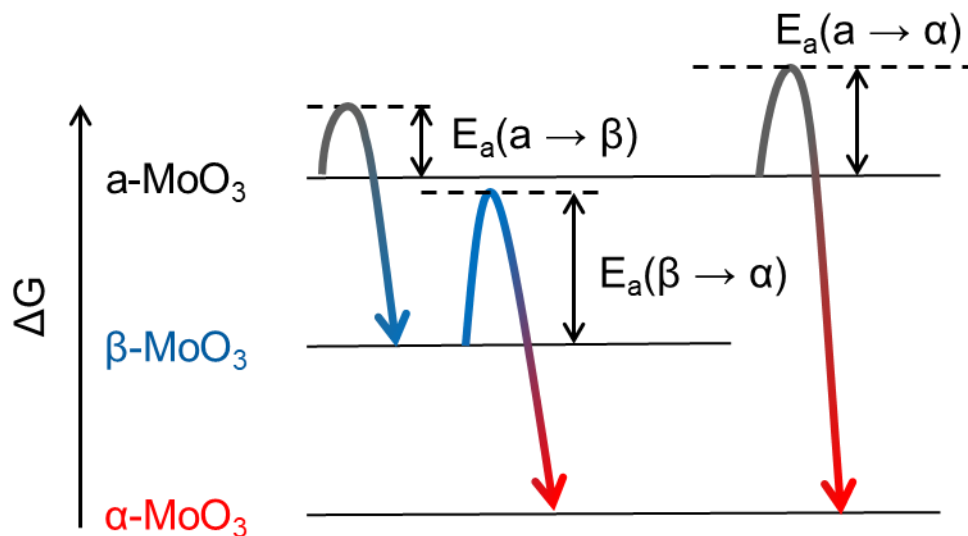


Figure S12 Energy diagram of the crystallization and phase transition in MoO₃. The diagram illustrates the activation energy of crystallization of a -MoO₃ and phase transition from β -MoO₃ to α -MoO₃. The relation of the activation energy can be summarized as $E_a(a \rightarrow \beta) < E_a(a \rightarrow \alpha) < E_a(\beta \rightarrow \alpha)$.

Alt text: Schematic of the activation energy diagram for the crystallization of amorphous and phase transition from β phase to α phases.

