Supplementary Information

Deposition of phosphorus-incorporated layered carbon nitride films and their electronic and magnetic properties

Natsuko Kurimoto[1], *Noriyuki Urakami*[1, 2, a)], *Yoshio Hashimoto*[1, 2], *and Xiaoxi Liu*[1, 2]

[1] *Department of Electrical and Computer Engineering,*

Shinshu University, 4-17-1 Wakasato, Nagano, 380-8553, Japan

[2] *Research Initiative for Supra-Materials,*

Shinshu University, 4-17-1 Wakasato, Nagano 380-8533, Japan

[a)] Author to whom correspondence should be addressed: urakami@shinshu-u.ac.jp

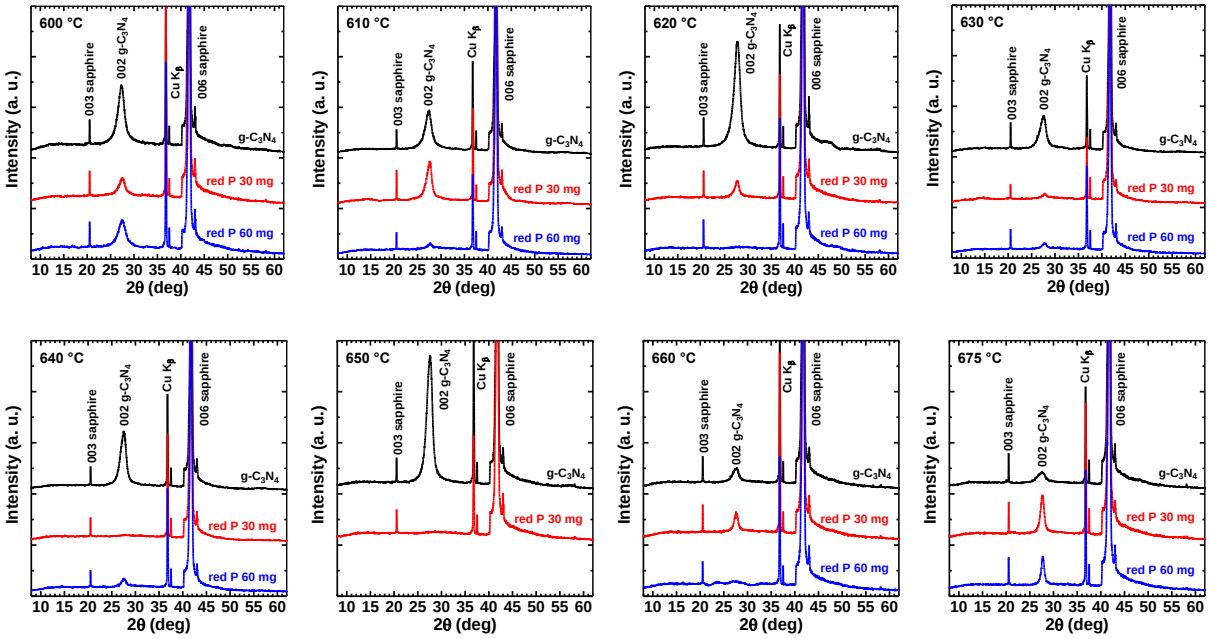


Figure S1. X-ray diffraction profiles for unintentionally doped and phosphorus (P)-incorporated g-C_3N_4 films fabricated at various deposition temperatures. The diffraction peaks at 37.5 ° correspond to CuKβ line.

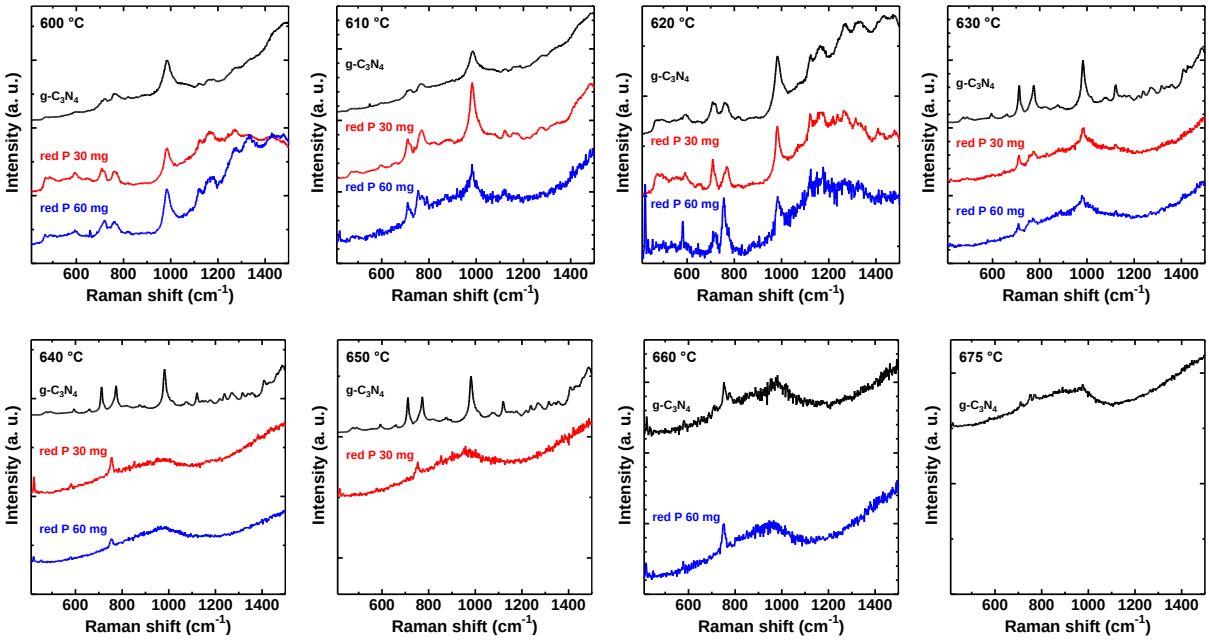
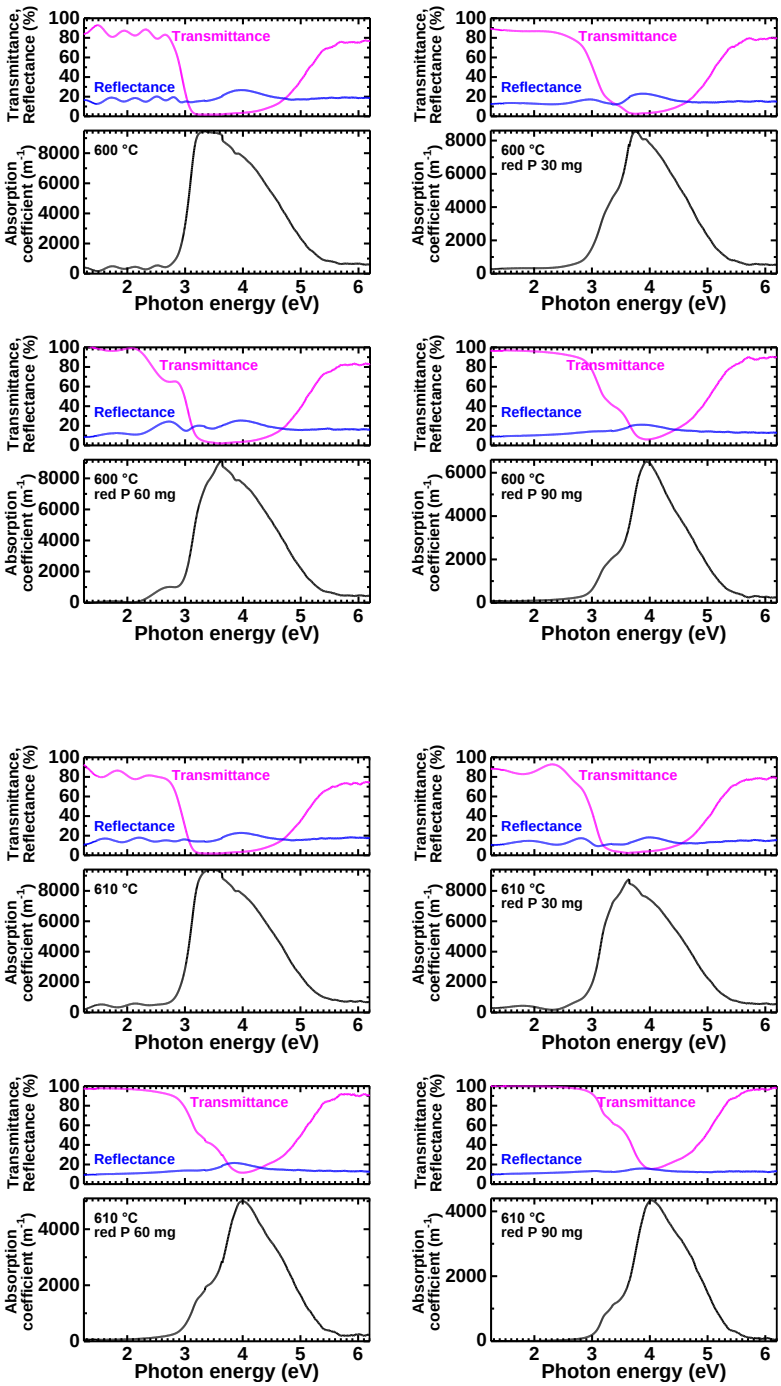
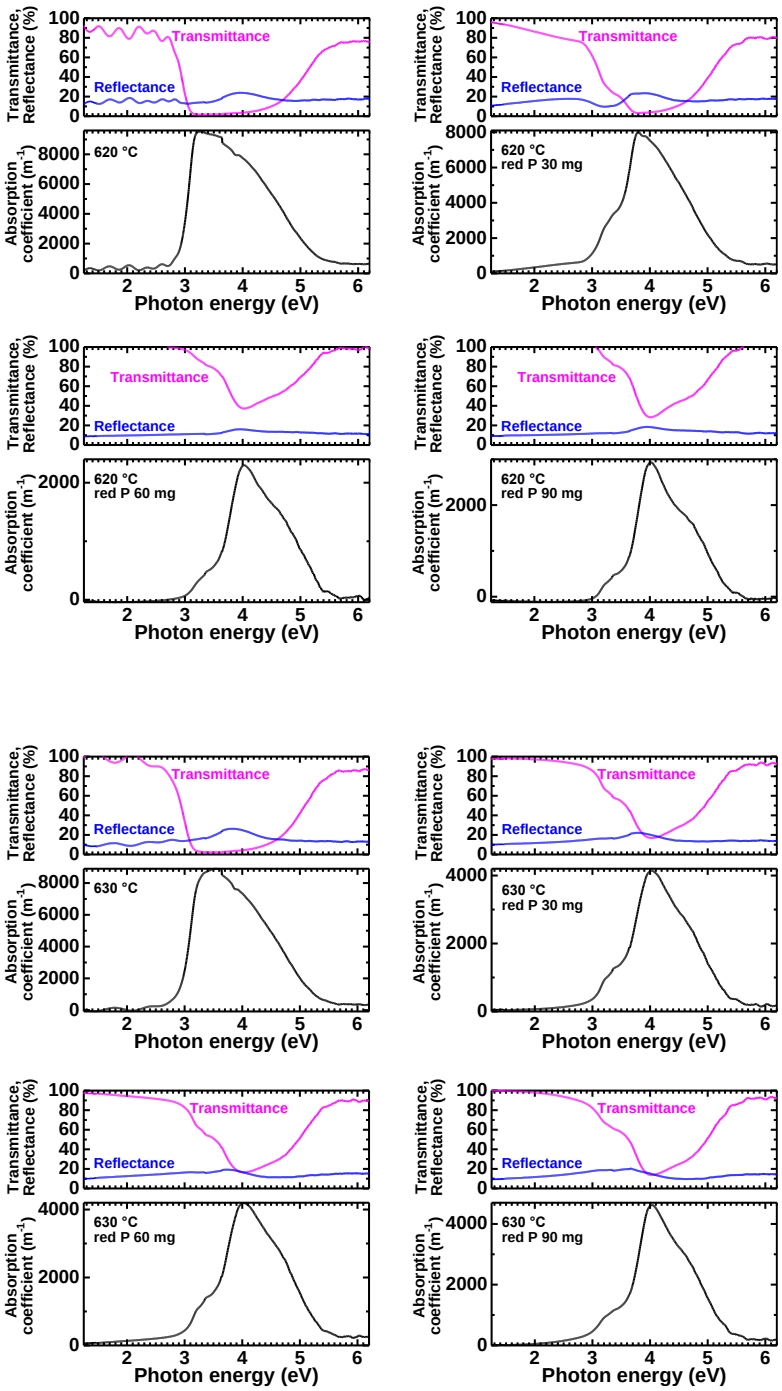
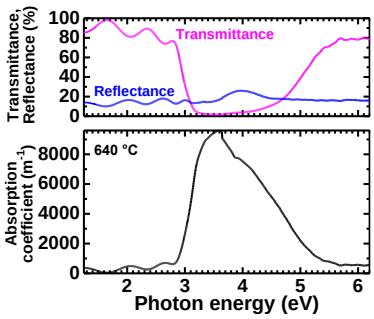
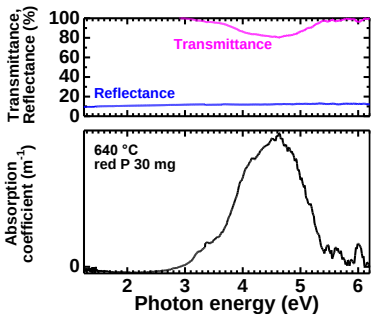


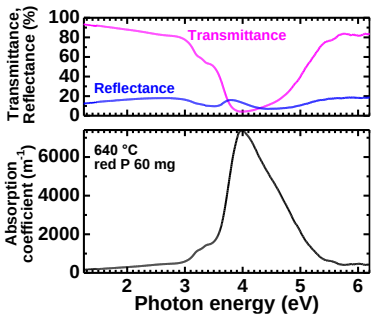
Figure S2. **Raman spectra for unintentionally doped and P-incorporated g-C_3N_4 films fabricated at various deposition temperatures.**

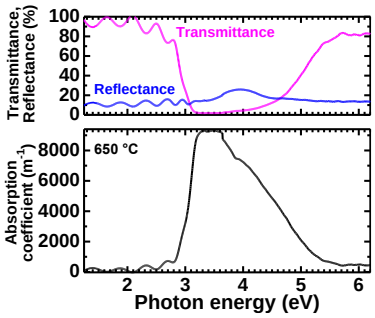

Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
600 °C
600 °C red P 30 mg
600 °C red P 60 mg
600 °C red P 90 mg
610 °C
610 °C red P 30 mg
610 °C red P 60 mg
610 °C red P 90 mg
Photon energy (eV)

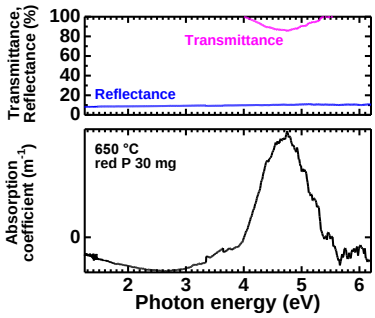

Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
620 °C
620 °C red P 30 mg
620 °C red P 60 mg
620 °C red P 90 mg
630 °C
630 °C red P 30 mg
630 °C red P 60 mg
630 °C red P 90 mg
Photon energy (eV)


Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
640 °C
Photon energy (eV)


Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
640 °C
red P 30 mg
Photon energy (eV)


Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
640 °C
red P 60 mg
Photon energy (eV)


Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
650 °C
Photon energy (eV)


Transmittance, Reflectance (%)
Transmittance
Reflectance
Absorption coefficient (m-1)
650 °C
red P 30 mg
Photon energy (eV)

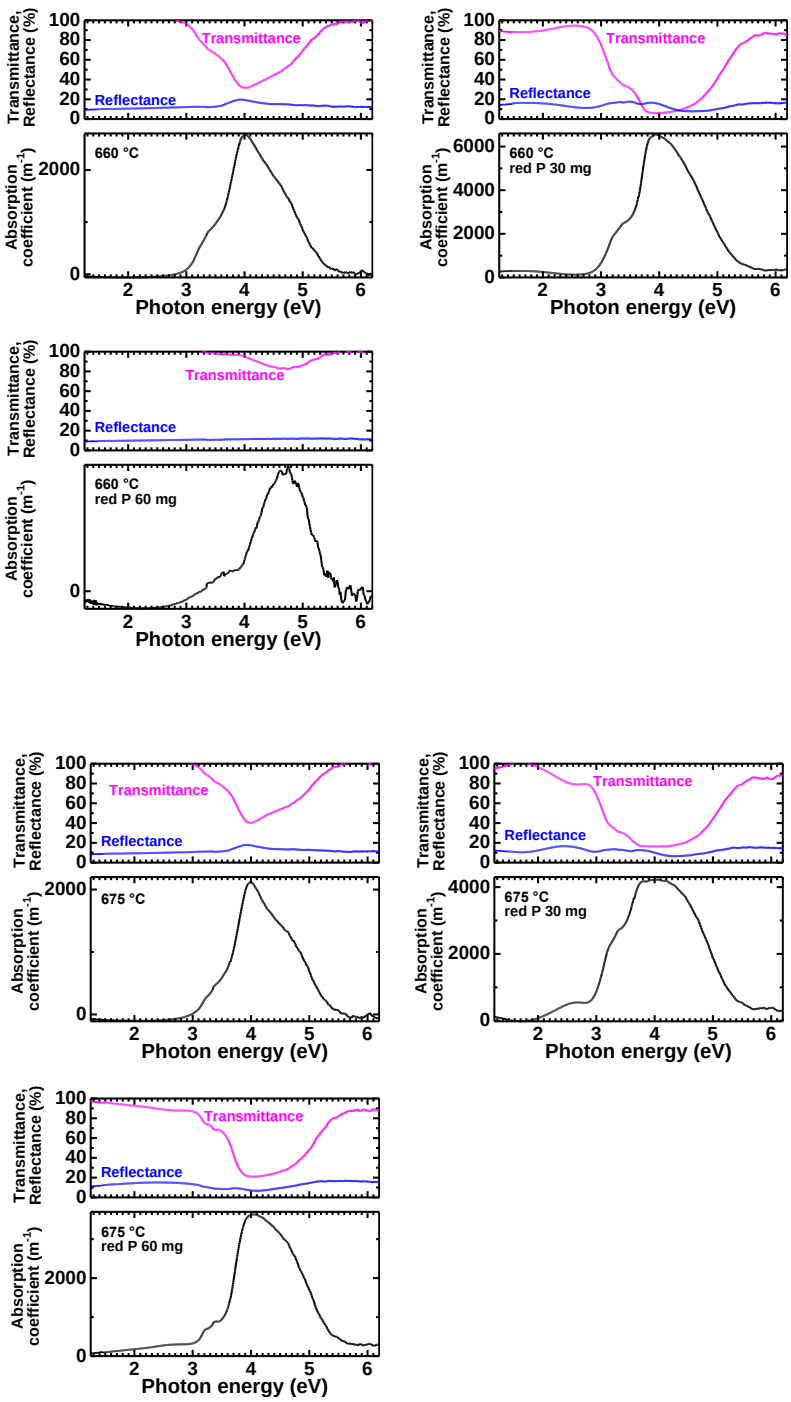
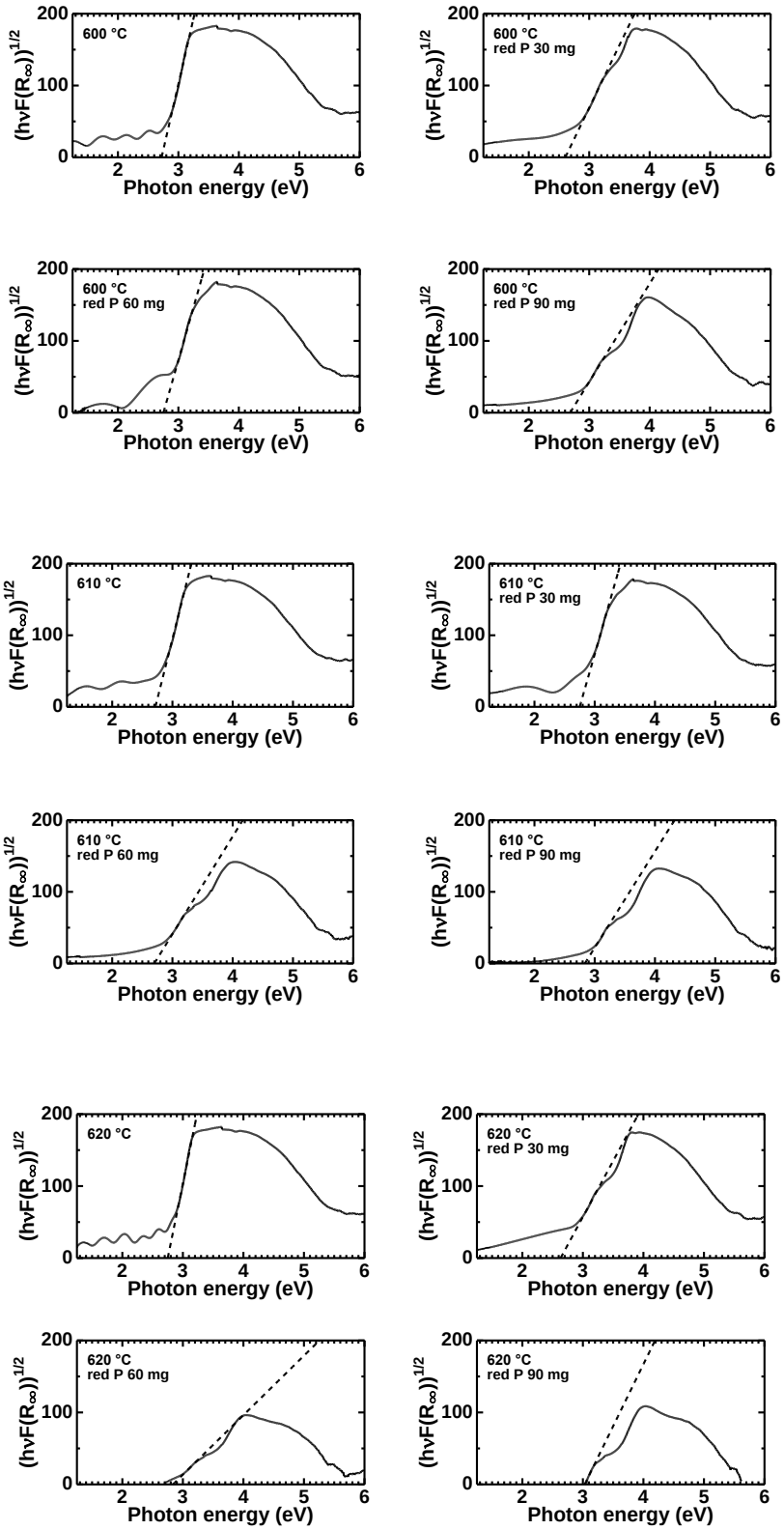
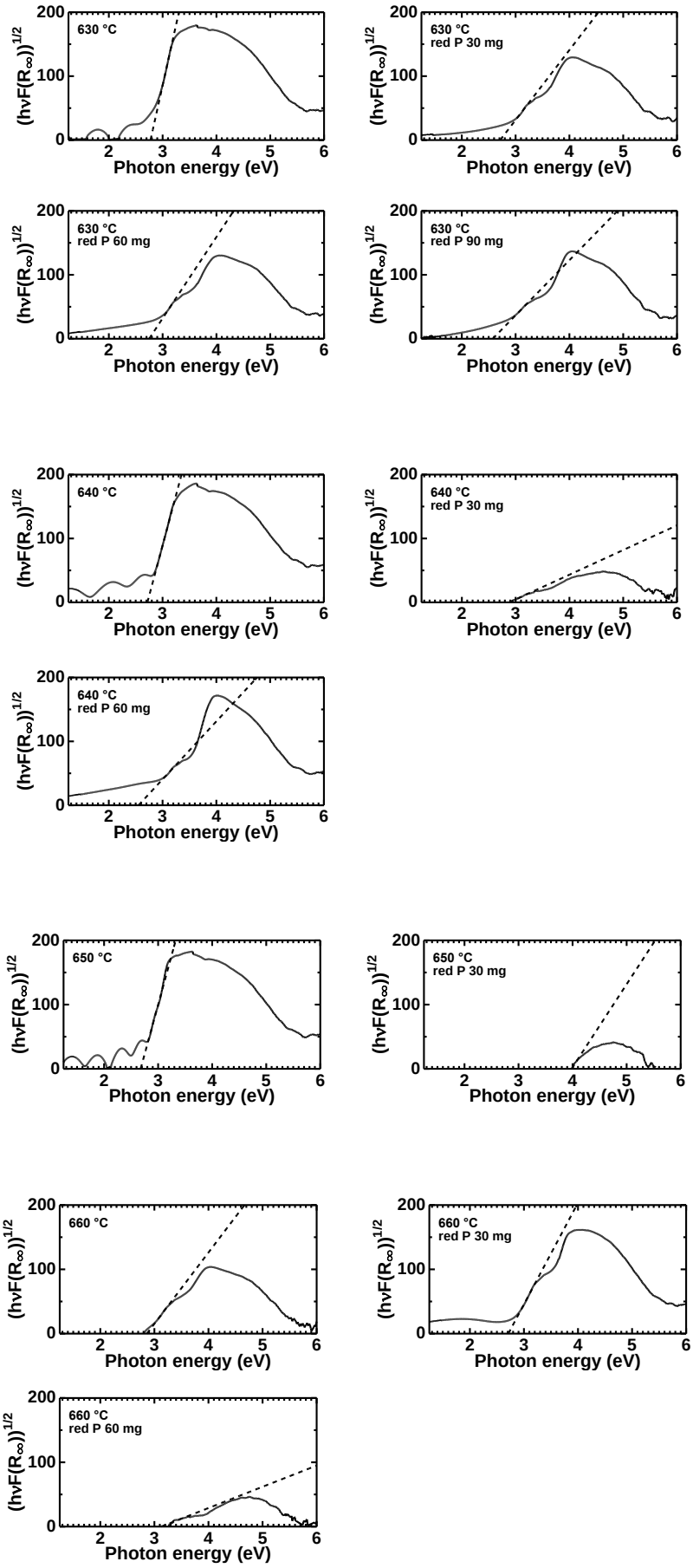


Figure S3. **Optical absorption coefficient for g-C_3N_4 films estimated using transmittance and reflectance spectroscopy. (Upper) Transmittance and reflectance spectra and (lower) absorption coefficient depending on photon energy.**


600 °C
600 °C
red P 30 mg
600 °C
red P 60 mg
600 °C
red P 90 mg
610 °C
610 °C
red P 30 mg
610 °C
red P 60 mg
610 °C
red P 90 mg
620 °C
620 °C
red P 30 mg
620 °C
red P 60 mg
620 °C
red P 90 mg
(hvF(R∞))^1/2
Photon energy (eV)
0
100
200
2
3
4
5
6


630 °C
630 °C
red P 30 mg
630 °C
red P 60 mg
630 °C
red P 90 mg
640 °C
640 °C
red P 30 mg
640 °C
red P 60 mg
650 °C
650 °C
red P 30 mg
660 °C
660 °C
red P 30 mg
660 °C
red P 60 mg
(hνF(R∞))^1/2
Photon energy (eV)
0
100
200
2
3
4
5
6

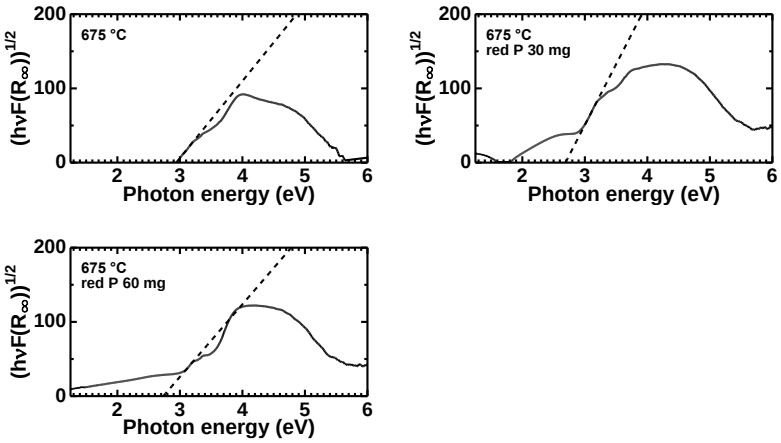
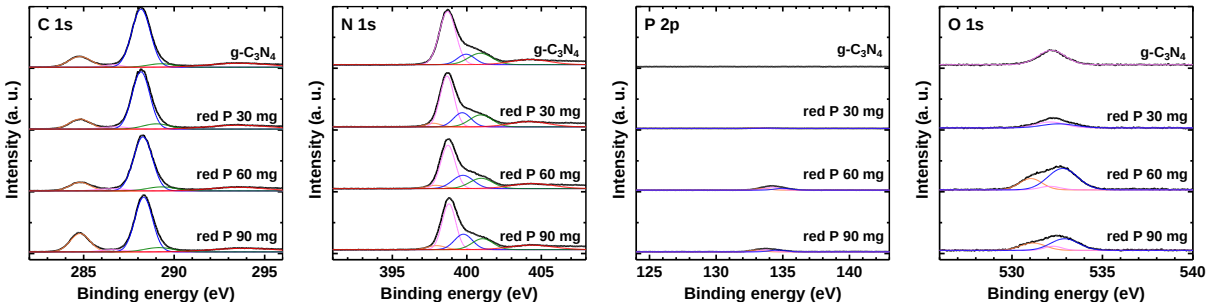
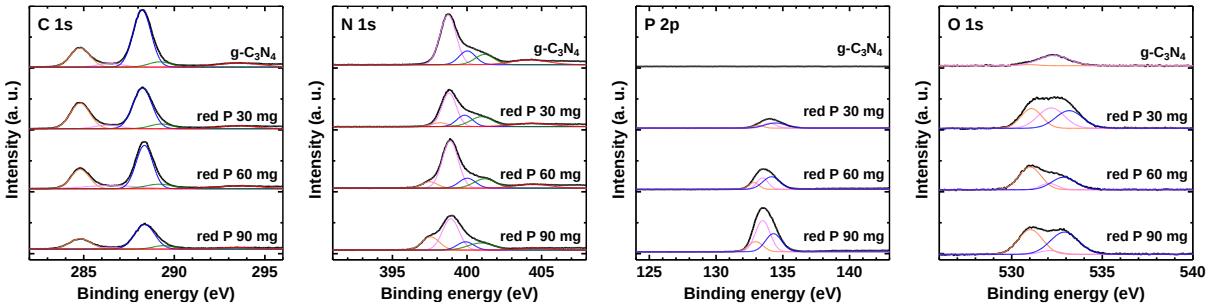


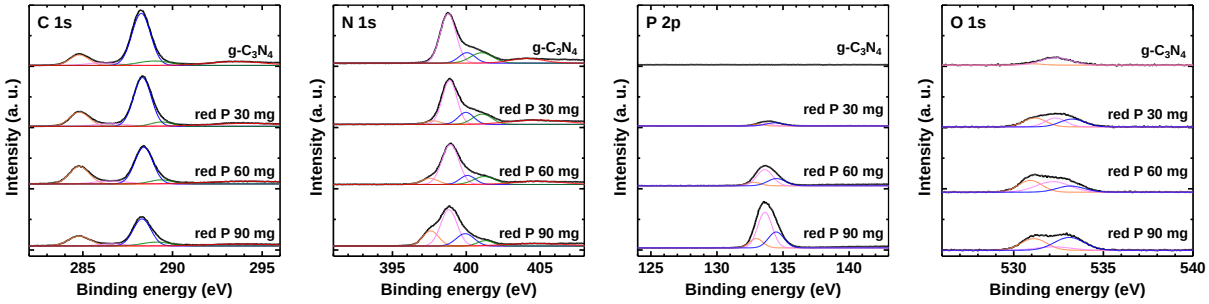
Figure S4. **Kubelka-Munk plots of g-C_3N_4 films, which were fitted to obtain the absorption coefficient as a function of photon energy.**



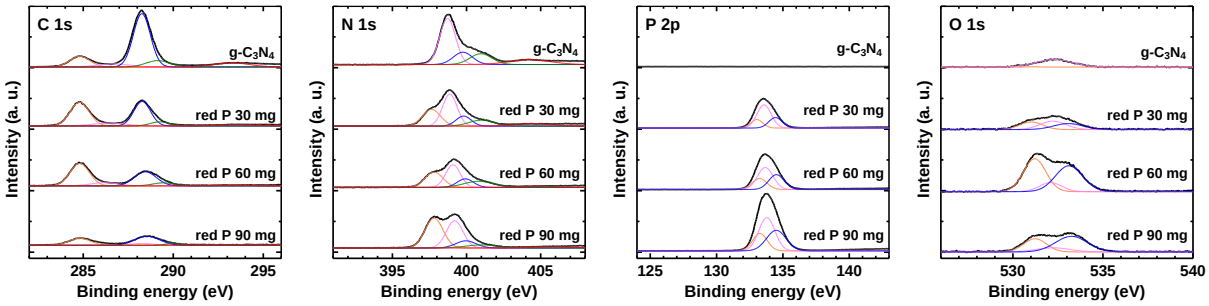
600 °C



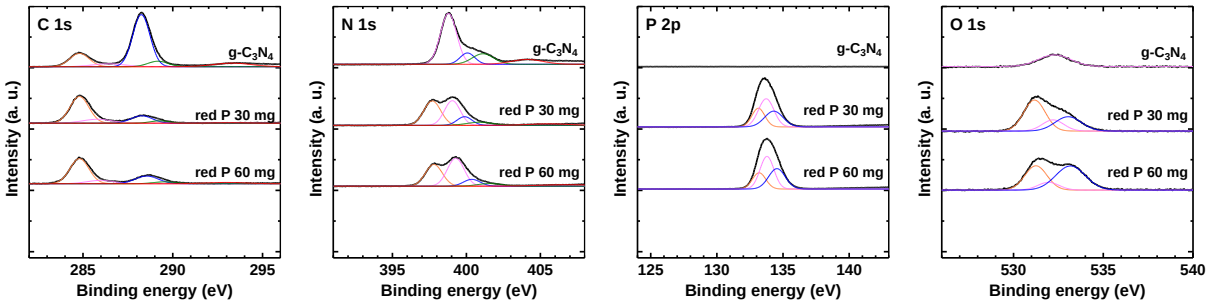
610 °C



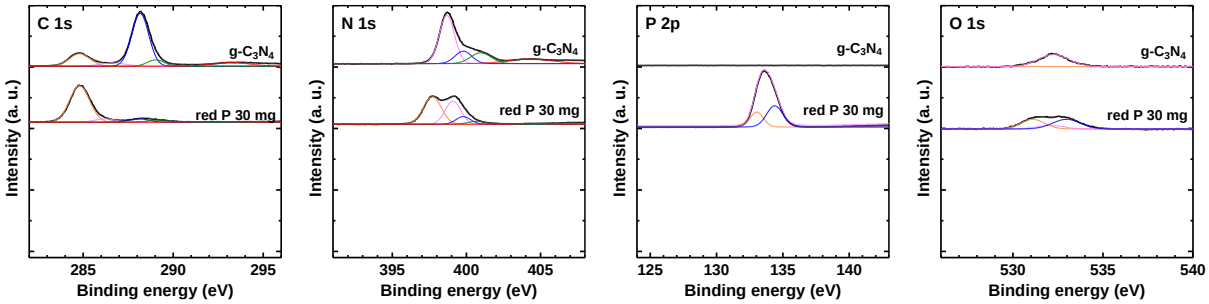
620 °C



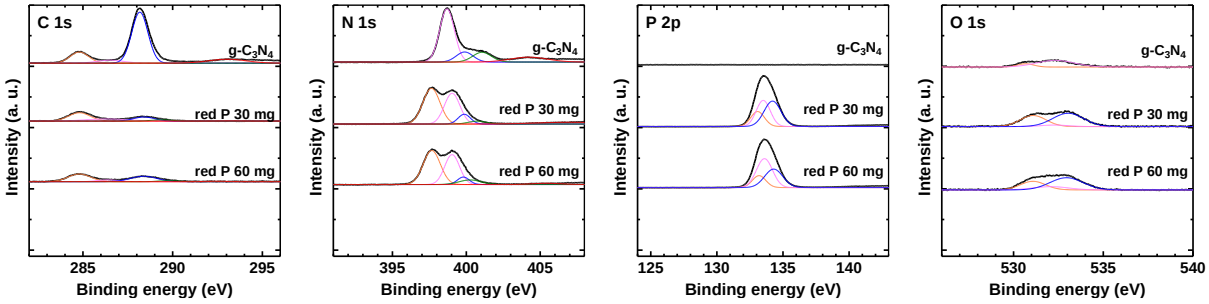
630 °C



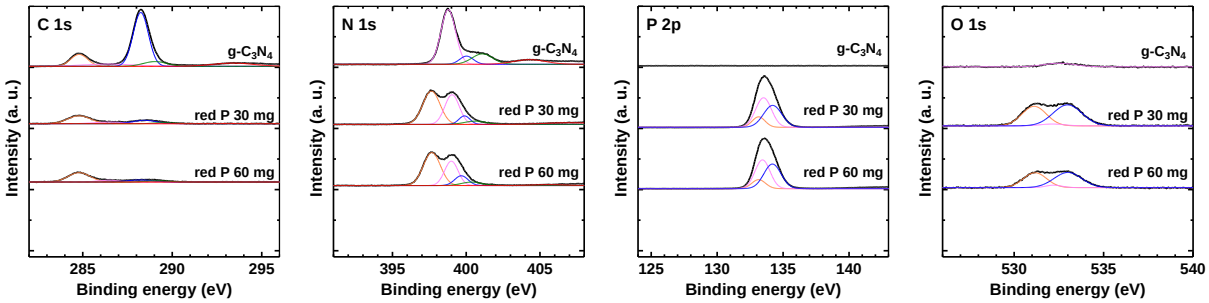
640 °C



650 °C

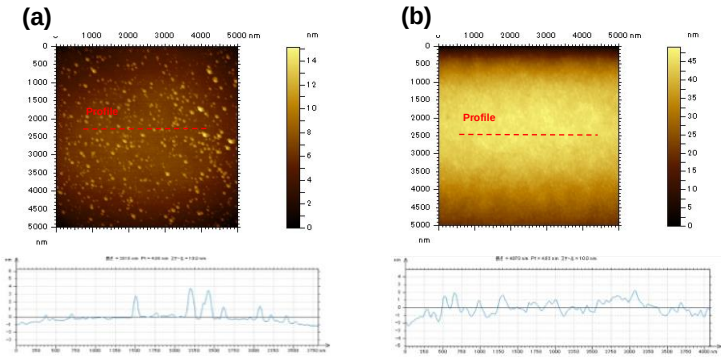


660 °C



675 °C

Figure S5. **C 1s, N 1s, P 2p, and O 1s core signals from the XPS analysis of g-C_3N_4 films.**



	(nm)
Sa	1.35
Sq	1.71
Sz	15.2

	(nm)
Sa	8.14
Sq	9.84
Sz	49.0

Figure **S6. AFM images, profiles and height parameters of (a) the unintentionally doped and (b) P-incorporated g-C_3N_4 films on the c-plane sapphire substrate. The g-C_3N_4 films were deposited at 610 °C with the amounts of P source of 60 mg. Height parameters of Sa, Sq and Sz correspond to the surface roughness (arithmetic mean height), RMS (root mean square) height and maximum height.**

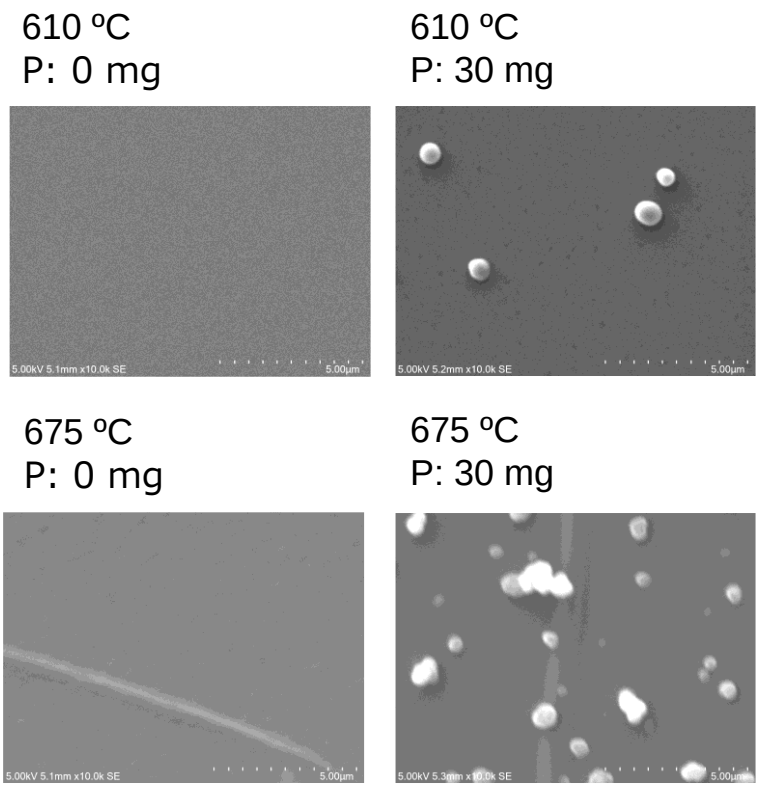


***Figure* S7. Surface SEM images of g-C_3N_4 films without and with the P incorporation deposited at 610 and 675 °C.**

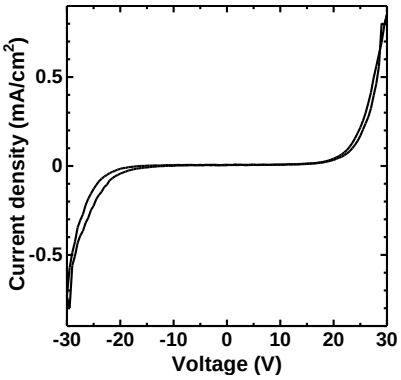


***Figure S8.* Current density – voltage characteristic along the out-of-plane direction for the g-C_3N_4 film with high electric resistivity. A space charge limited conduction is observed, as reported previously.[R1, R2]**

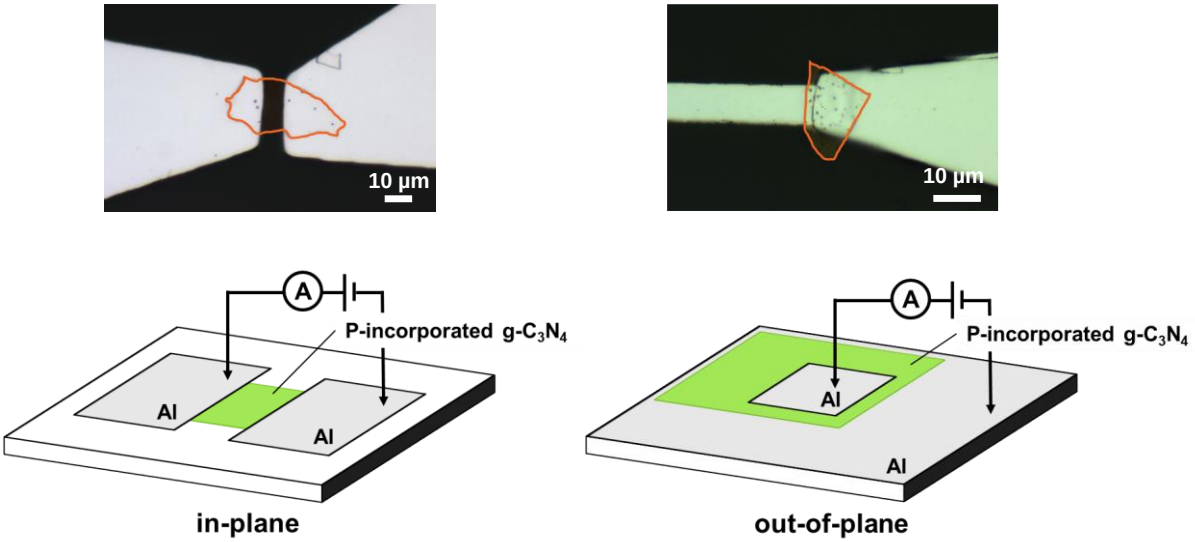


Figure S9. **Setups of the transport measurement for the (left) in-plane and (fight) out-of-plane directions.**

References

R1. P. C. Patra and Y. N. Mohapatra, "Dielectric constant of thin film graphitic carbon nitride (g- C_3N_4) and double dielectric Al_2O_3/g-C_3N_4," *Appl. Phys. Lett.* **118**, 103501 (2021).

R2. K. Higuchi, M. Tachibana, N. Urakami, and Y. Hashimoto, "Layered carbon nitride films deposited under an oxygen-containing atmosphere and their electronic properties", *AIP Adv.* **14**, 025047 (2024).