# Combining single source chemical vapour deposition precursors to explore the phase space of titanium oxynitride thin films

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**ABSTRACT:**

In this paper we report on a novel chemical vapour deposition approach to the formation and control of composition of mixed anion materials, as applied to titanium oxynitride thin films. The method used is the aerosol assisted chemical vapour deposition (AACVD) of a mixture of single source precursors. To explore the titanium-oxygen-nitrogen system the single source precursors selected were tetrakis(dimethylamido) titanium and titanium tetraisopropoxide which individually are precursors to thin films of titanium nitride and titanium dioxide respectively. However, by combining these precursors in specific ratios in a series of AACVD reactions at 400 °C, we are able to deposit thin films of titanium oxynitride with three different structure types and a wide range of compositions. Using this precursor system we can observe films of nitrogen doped anatase, with 25% anion doping of nitrogen; a new composition of pseudobrookite titanium oxynitride with a composition of $Ti_3O_{3.5}N_{1.5}$, identified as being a UV photocatalyst; and rock-salt titanium oxynitride in the range $TiO_{0.41}N_{0.59}$ to $TiO_{0.05}N_{0.95}$. The films were characterised using GIXRD, WDX and UV-vis spectroscopy, and in the case of the pseudobrookite films, assessed for photocatalytic activity. This work shows that a so called dual single-source CVD approach is an effective method for the deposition of ternary mixed anion ceramic films through simple control of the ratio of the precursors, while keeping all other experimental parameters constant.

# SUPPORTING INFORMATION

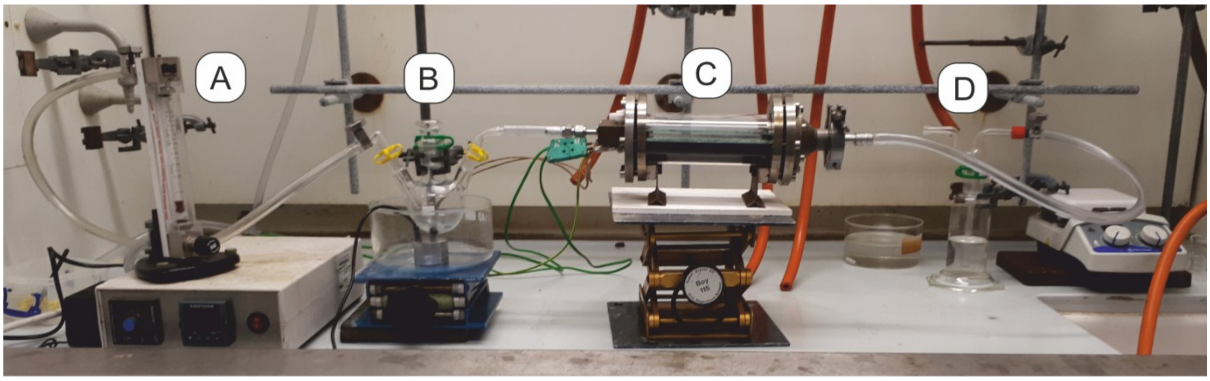

Figure S1. Photograph of the aerosol assisted chemical vapour deposition reactor set up. (A) Rotometer which allows controlled flow of argon carrier gas into the system. (B) Precursor supply point, consisting of a 100 ml three-necked round bottom flask, containing the precursor. This is suspended above a piezoelectric humidifier which generatres an aerosl mist of the precursor solution, which can then be carried to the reactor by the incoming gas. (C) Main reactor composed of a silica tube, with carbon block insert and Watlow heater cartrides. (D). Silicone filled bubbler, preventing back diffusion of air into the reactor.

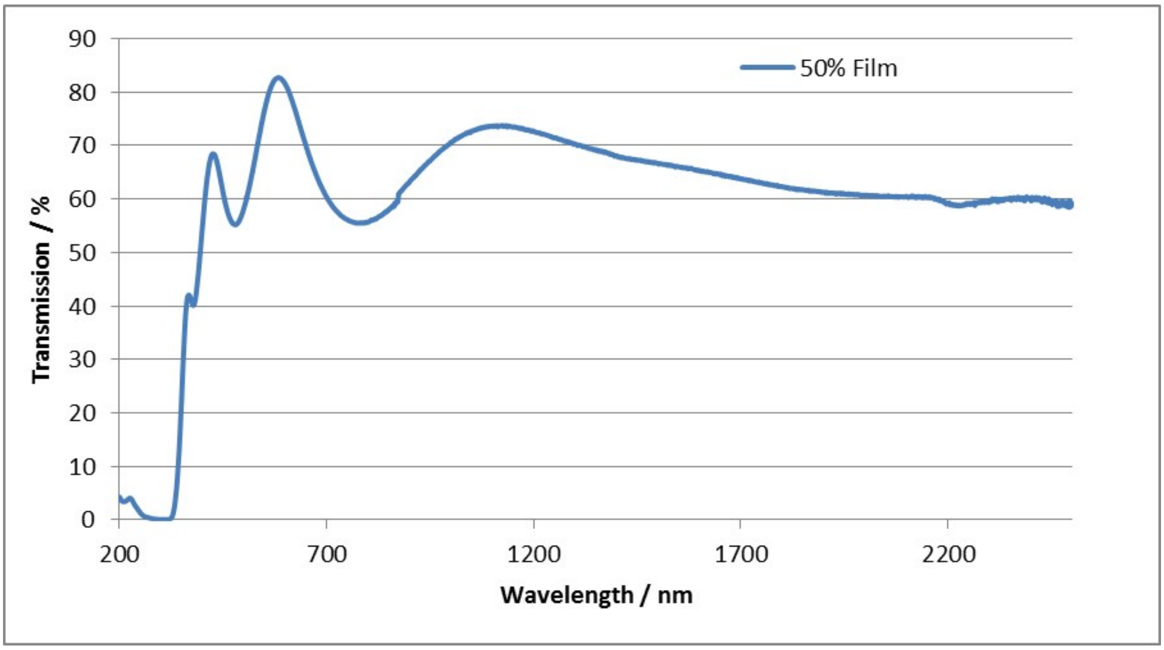

Figure S2. Transmission spectra of film formed using 50% TDMAT and 50% TTIP

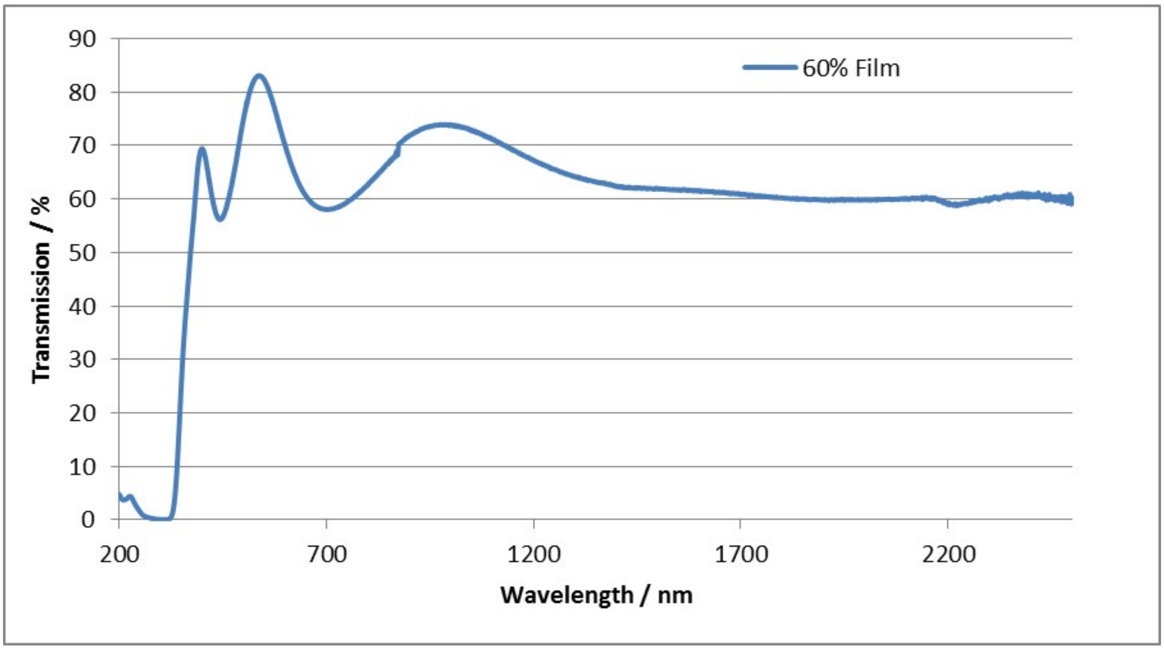

Figure S3. Transmission spectra of film formed using 60% TDMAT and 40% TTIP

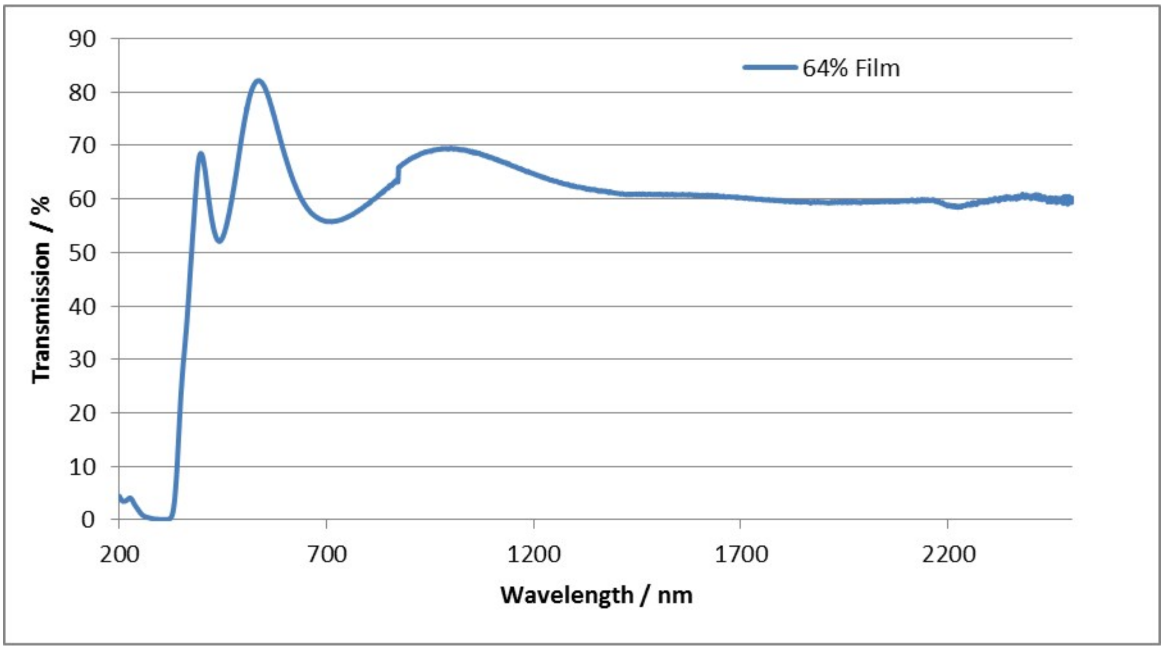

Figure S4. Transmission spectra of film formed using 64% TDMAT and 36% TTIP

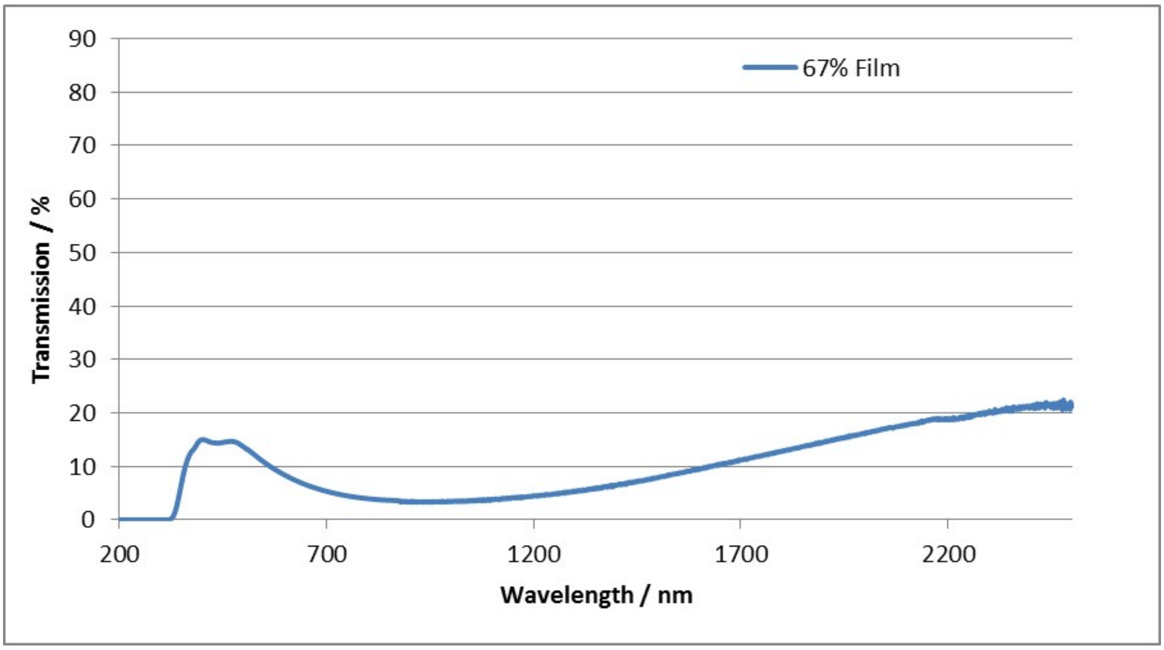

Figure S5. Transmission spectra of film formed using 67% TDMAT and 33% TTIP

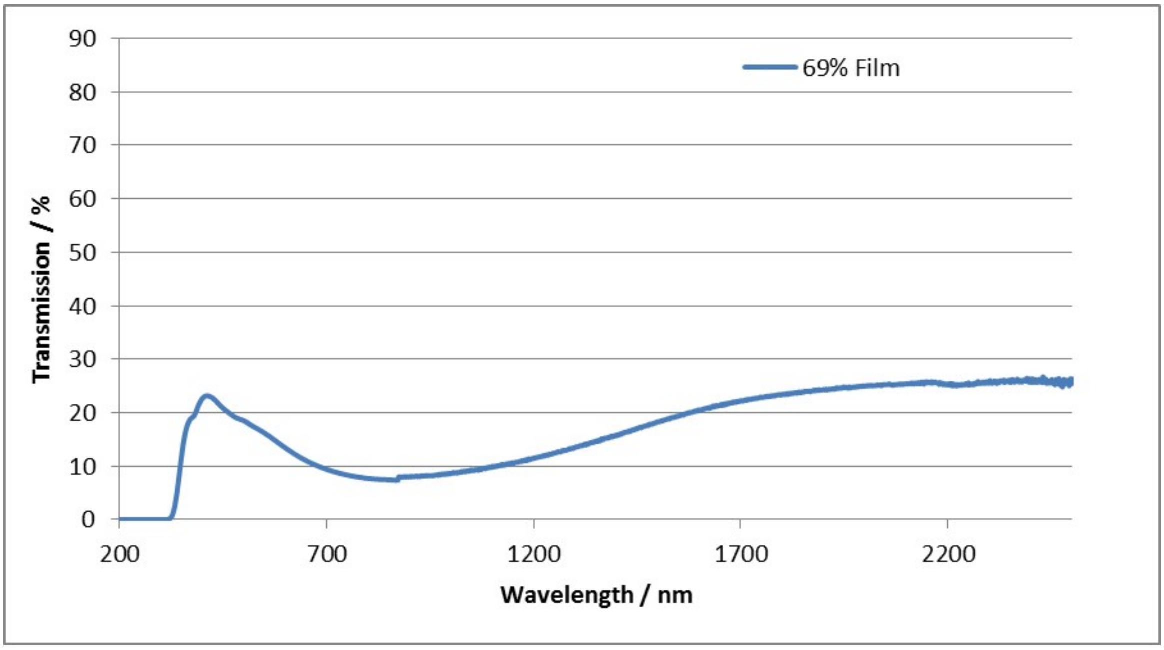

Figure S6. Transmission spectra of film formed using 69% TDMAT and 31% TTIP

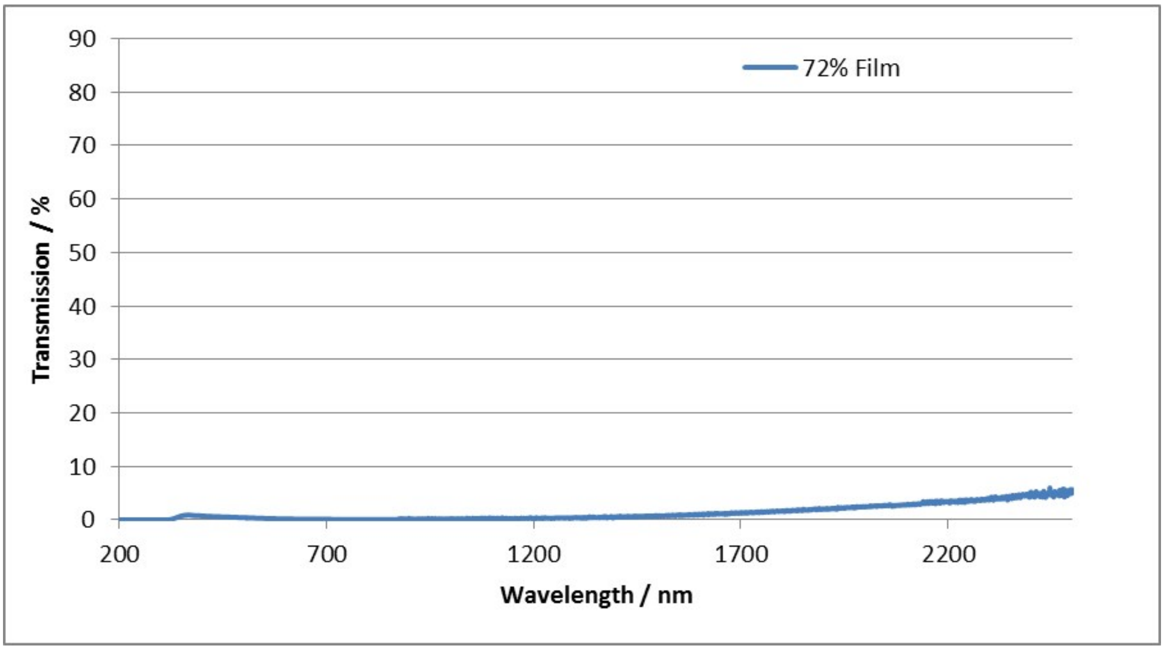

Figure S7. Transmission spectra of film formed using 72% TDMAT and 28% TTIP

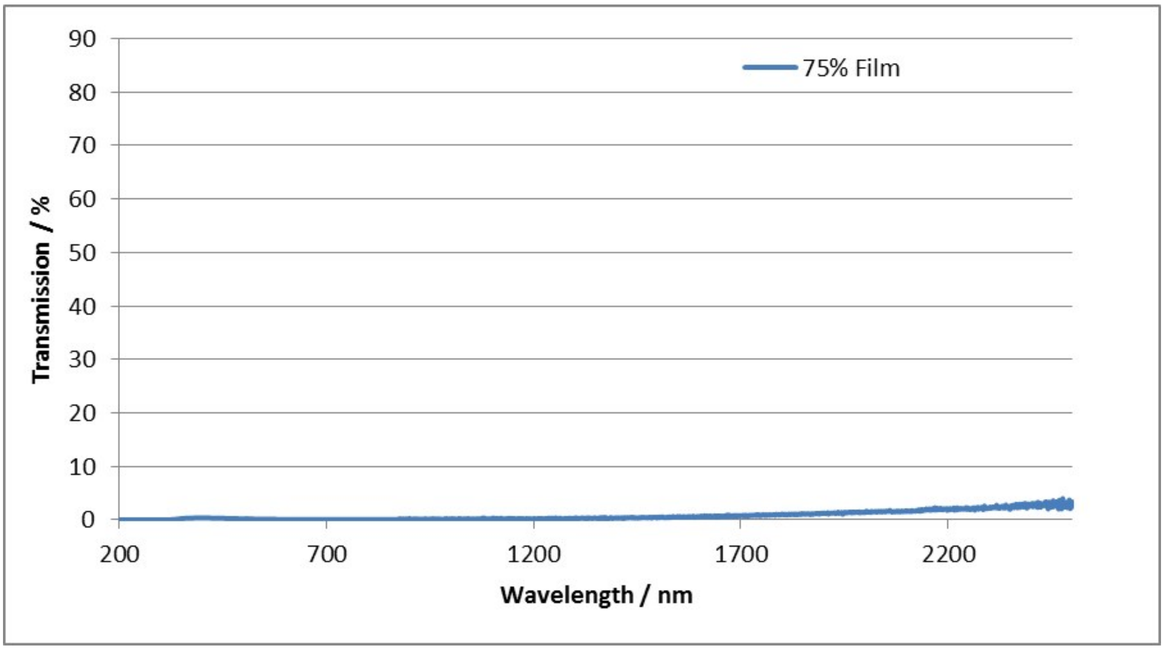

Figure S8. Transmission spectra of film formed using 75% TDMAT and 25% TTIP

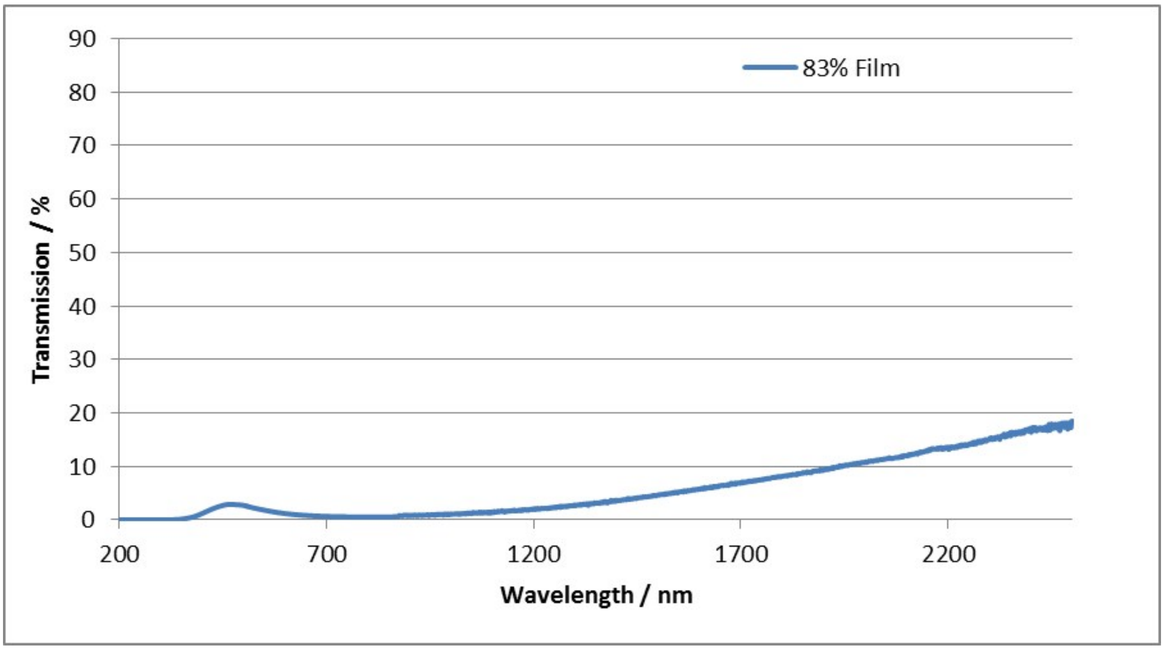

Figure S9. Transmission spectra of film formed using 83% TDMAT and 17% TTIP

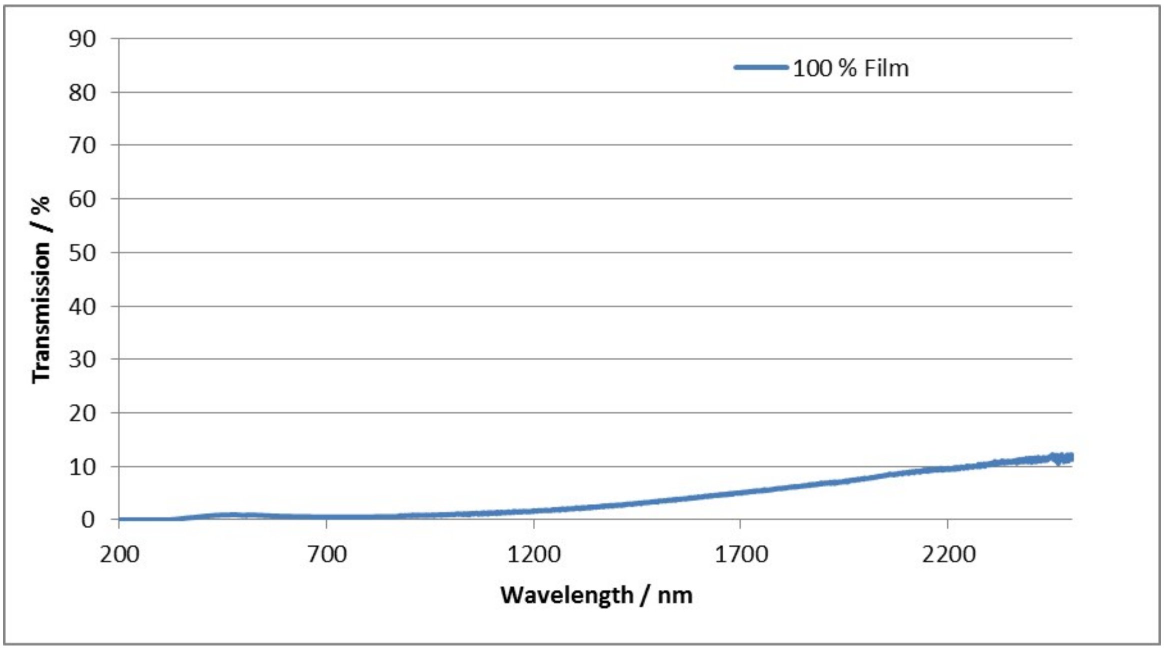

Figure S10. Transmission spectra of film formed using 100% TDMAT

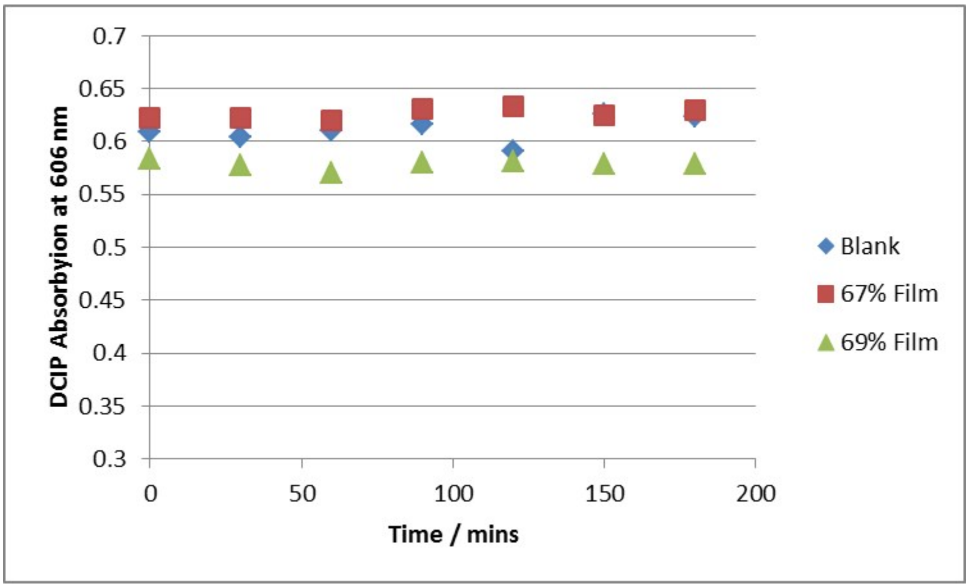

Figure S11. Plot of the absorption of DCIP dye as a function of time, for the photocatalytic experiments conducted using visible light

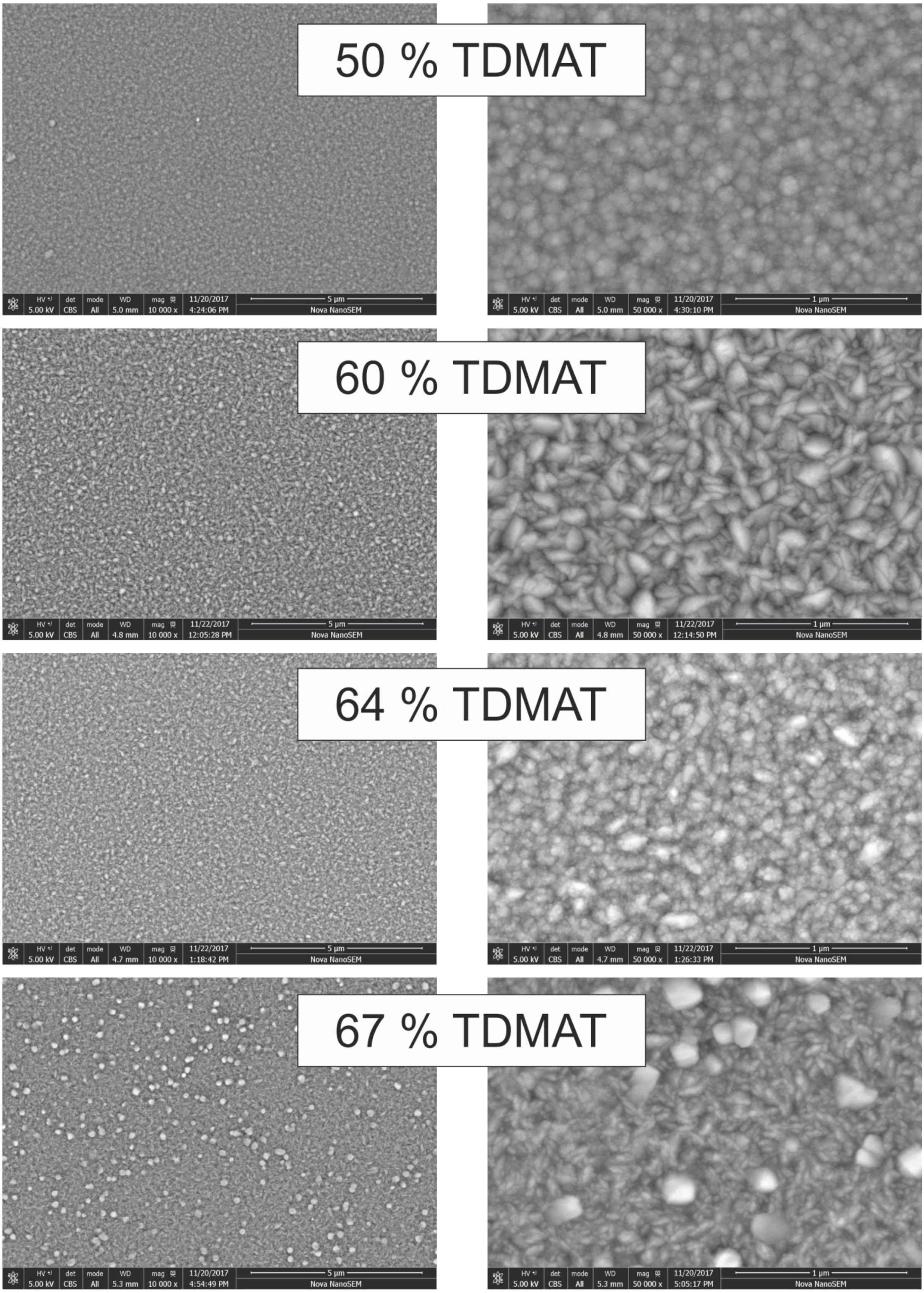

Figure S12. SEM images of the film surfaces

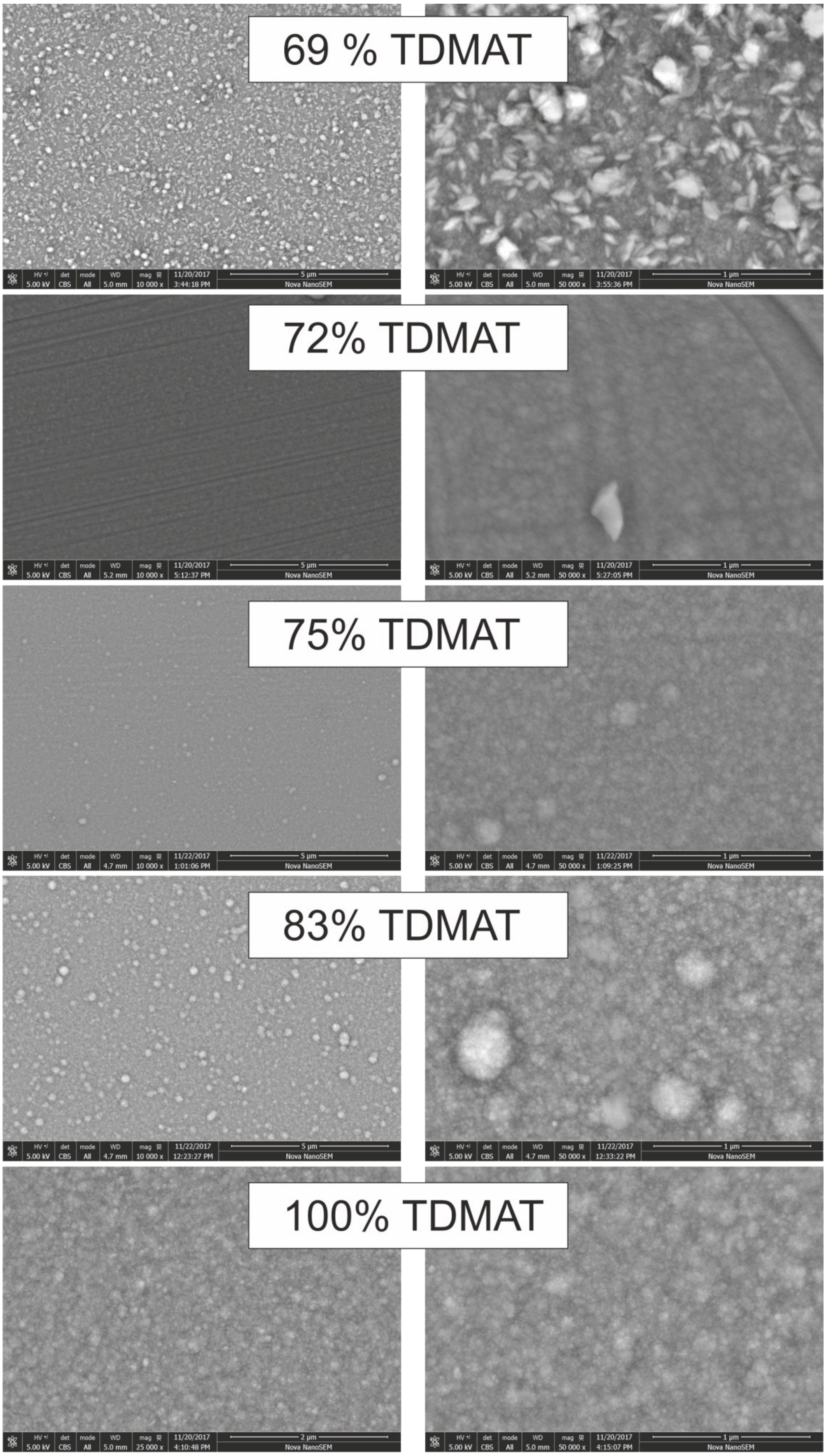

Figure S13. SEM images of the film surfaces

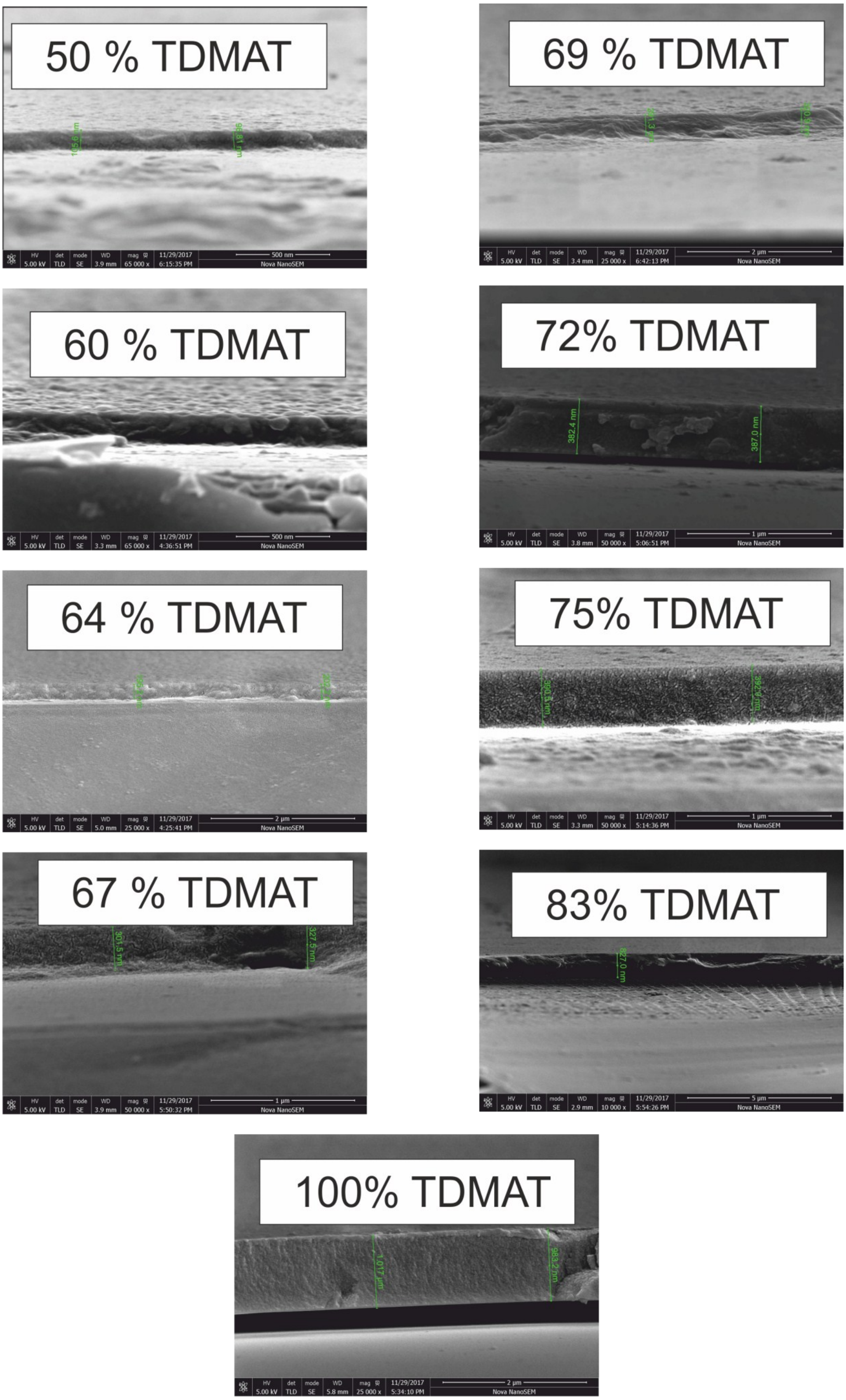

Figure S14. Side-on SEM images of the films