Supplementary information for

High-throughput roll-to-roll fabrication of flexible thermochromic coatings for smart windows with VO_2 nanoparticles

Youngkwang Kim[1], Sangbae Yu[1], Jaeseoung Park[1], Daseob Yoon[1], Amir Masoud Dayaghi[1], KunJoong Kim[1], Jin Soo Ahn[2], and Junwoo Son[1,*]

[1] Department of Materials Science and Engineering, Pohang University of Science and Technology (POSTECH), Pohang 37673, Republic of Korea

[2] Research Institute of Industrial Science and Technology (RIST), Pohang 37679, Republic of Korea

[*] jwson@postech.ac.kr

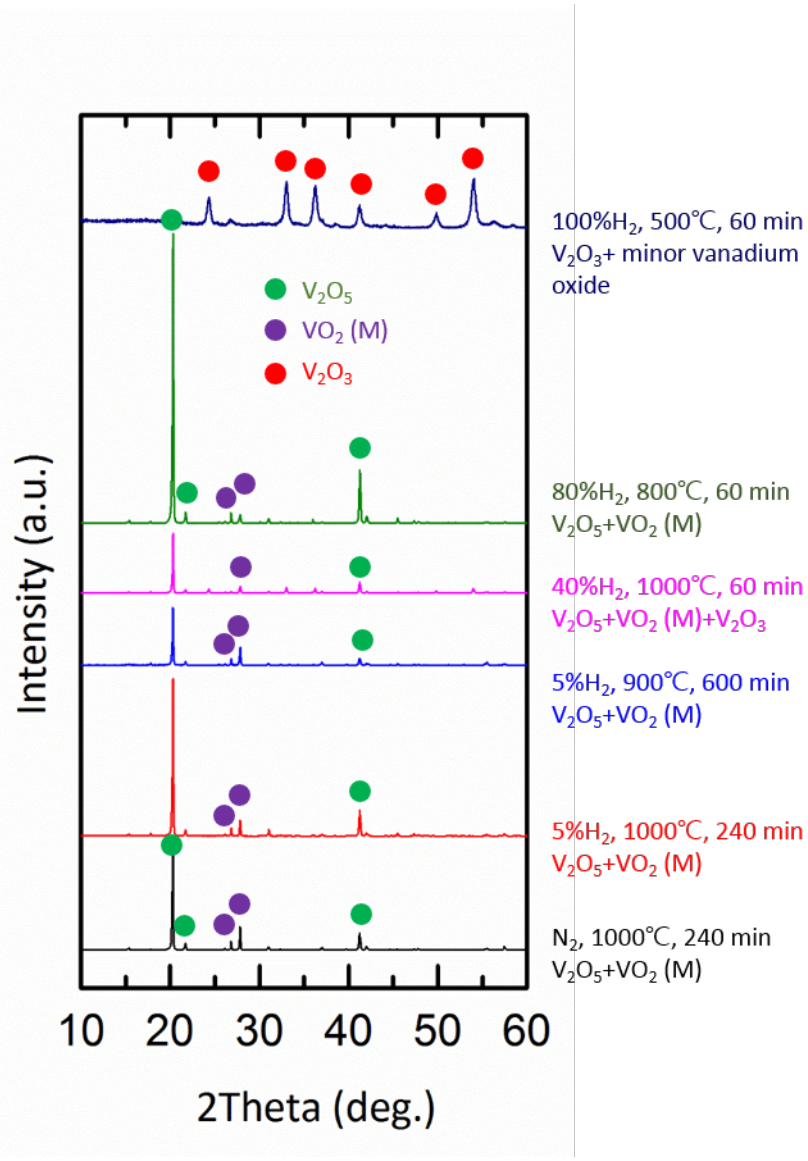

Figure S1 | The difficulty in synthesis of uniform VO_2 particles from micron-sized V_2O_5 particles. V_2O_5 and other vanadium sub-oxides co-exist after reduction of V_2O_5 micro-sized powders, probably because oxygen tends to be lost more easily from near the surfaces of particles than from their cores. This non-uniform reduction leads to uneven distribution of the amount of oxygen, and that of valence states of vanadium ions in the particles; Due to the complex stoichiometry of vanadium oxides, the thermodynamic condition for uniform phase transformation (V_2O_5 to VO_2) appears to be very difficult simply by controlling oxygen partial pressure during thermal treatment when micron-sized powders are used

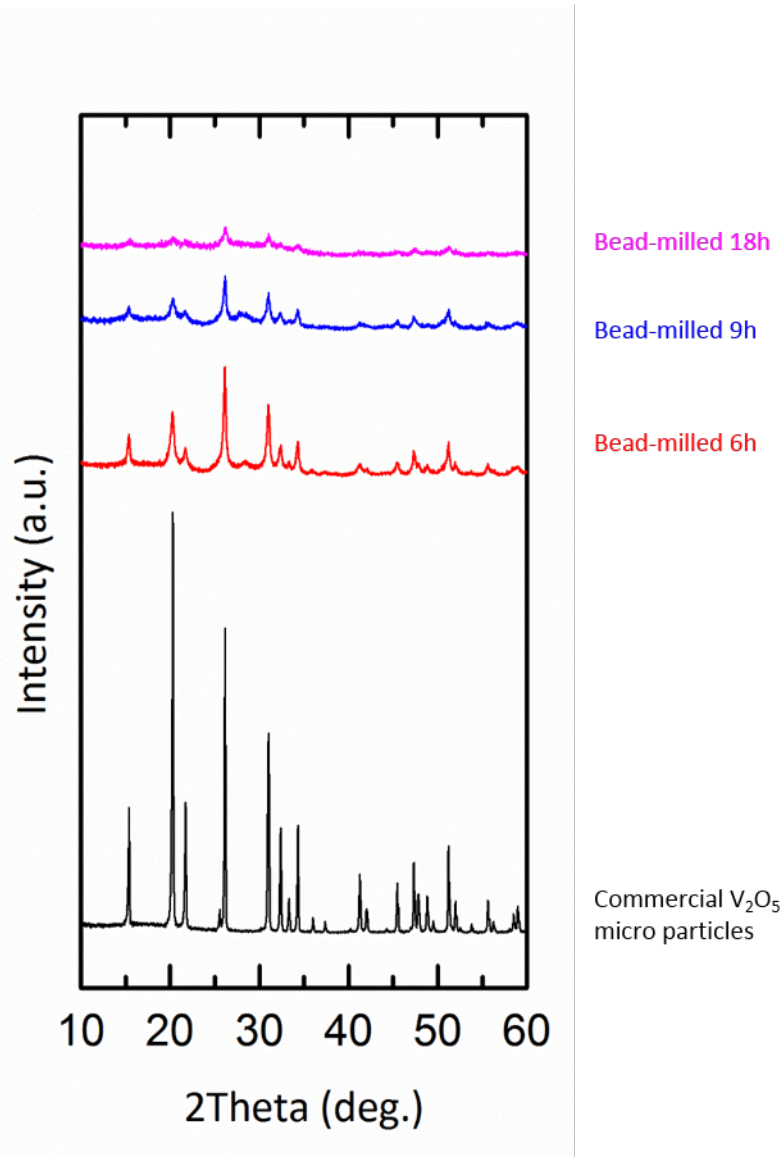

Figure S2 | The change of XRD diffraction in V2O5 particles as a function of bead milling time. The peak positions of x-ray diffraction (XRD) peaks for V_2O_5 phase did not move, but their width increased and their intensity decreased as bead-milling time increased; this trend indicates that milling decreased the size of V_2O_5 particles at the expense of their amorphization.

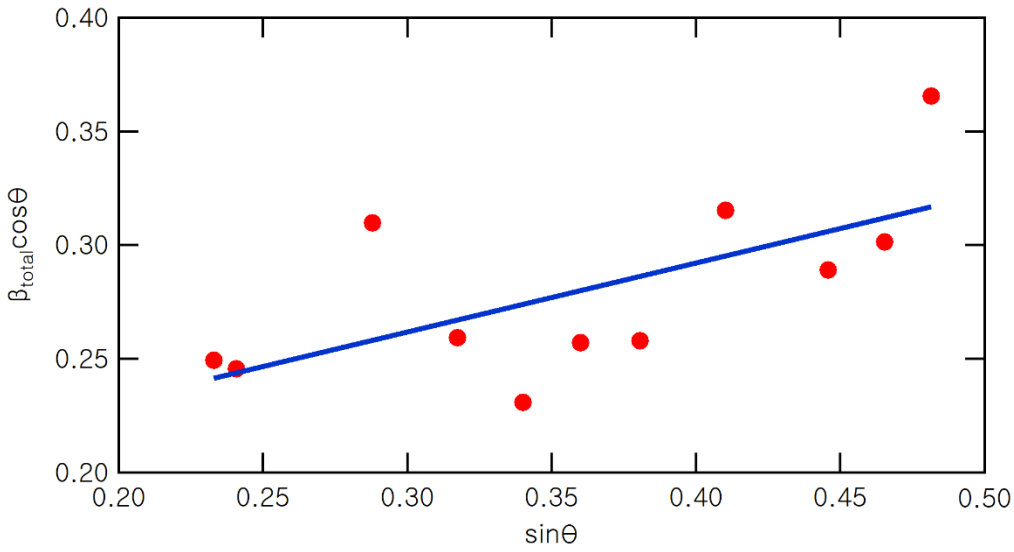

Figure S3| Williamsone Hall plot of our VO_2 NPs

It was usually known that crystallite size and strain are the two main sources related to peak broadening in XRD measurement. Williamsone Hall analysis considers these two main source to understand the measured peak broadening, and vice versa.

First of all, the relation between peak broadening and crystallite size, which is also known as Scherer equation, is

$$\beta_L = \frac{K\,\lambda}{L\cos\theta}$$

where β_L, K, λ, L, and θ are peak broadening, constant close to unity and often taken as 0.9, radiation wavelength, crystalline size, and Bragg angle, respectively.

Secondly, the relation between peak broadenning and inhomogeous strain has simple form of $\beta_\varepsilon = C_\varepsilon \tan\theta$, where β_ε, C_ε, θ are peak broadenging, constant close to 4 ~5, Bragg angle, respectively.

The Williamsone Hall analysis assumption is the total peak broadenning is simply the sum of β_L *and* β_ε. Using above equation then we get

$$\beta_{total} = \beta_L + \beta_\varepsilon = \frac{K\,\lambda}{L\cos\theta} + C_\varepsilon \tan\theta$$

$\beta_{total}\cos\theta = C_\varepsilon \sin\theta + \frac{K\,\lambda}{L}$, which is the same form of y = mx + c. Thus by plotting $\beta_{total}\cos\theta$ versus $\sin\theta$, we get the size factor from the intercept ($\frac{K\,\lambda}{L}$) and then we can obtain the crystallite size (L).

Below is the plot of $\beta_{total}\cos\theta$ and $\sin\theta$ value of each Bragg peak of our synthesized VO_2 NPs (**Fig. S3**).

The average size of VO_2 NPs was calculated to be 45.1 nm by using the intercept value ($\frac{K\,\lambda}{L}$), which was comparable to the average value measured by TEM image (~ 38 nm).