Supporting Information

Unravelling the Mechanism of Phase Fraction Modulation via Process Parameter Tuning and First-Principles Study for Enhanced TCR in VO_x-Based Uncooled Microbolometers

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Figure S1: The architecture of VO_x films deposited on $Si/SiO_2/SiN_x$ substrate.

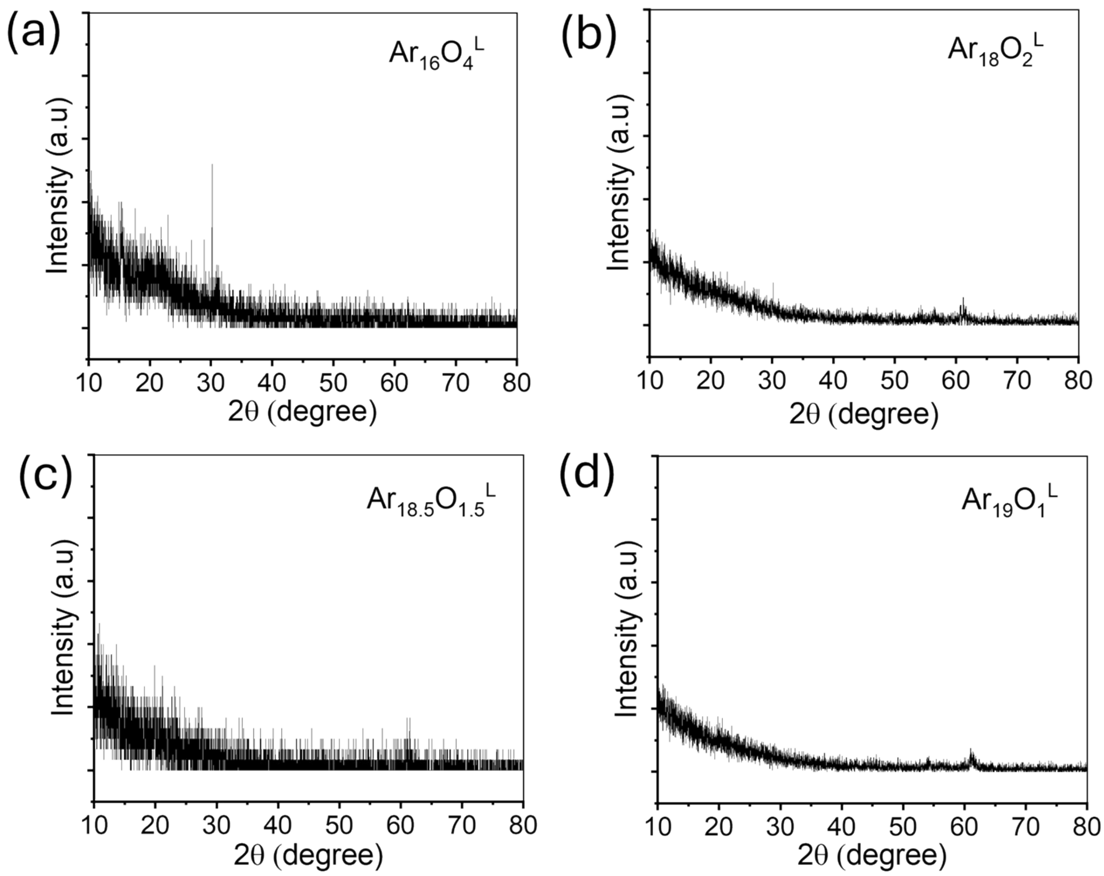

Figure S2: XRD pattern of films deposited at 250 °C

Table S1: Crystallite size of samples deposited at high temperature.

Sample no	Sample Name	Crystallite Size (nm)
1	$Ar_{19}O_1^H$	15.4
2	$Ar_{18.5}O_{1.5}^H$	17.1
3	$Ar_{18}O_2^H$	20.3
4	$Ar_{16}O_4^H$	34.9

Note: Although, we would like to mention that the compositional analysis through XPS is not an accurate technique as most of the surface of VO_x is highly oxidised to V_2O_5 due to ambient atmosphere exposure (Begara et al. Applied Surface Science, 2017). Also, to remove this oxide we can etch the surface but that changes the oxidation state as reported by Silversmit et al. Surface Science, Vol. 600, 2006. Thus, XPS is more suitable to understand the presence of a particular oxidation state, but compositional analysis through XPS might not give very accurate results and can only give a rough estimate.

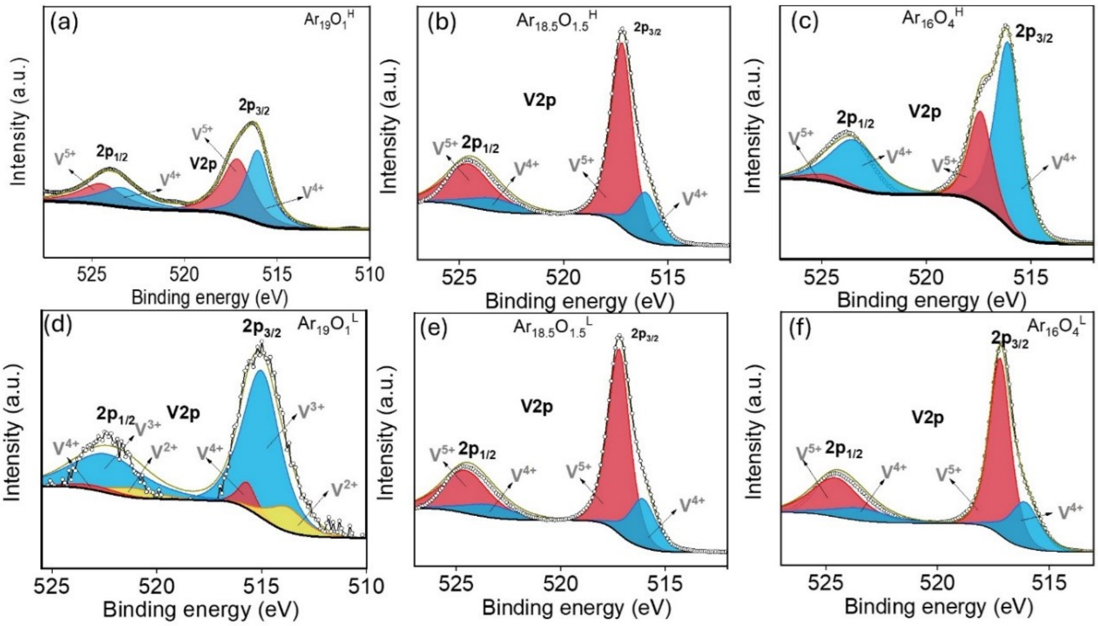

Figure S3: XPS spectra of samples (a) $Ar_{19}O_1^H$ and (b) $Ar_{18.5}O_{1.5}^H$ (c) $Ar_{16}O_4^H$ and (d) $Ar_{19}O_1^L$ (e) $Ar_{18.5}O_{1.5}^L$ and (F) $Ar_{16}O_4^L$

Table S2: Composition of films analysed from XPS date.

S. No.	Sample name	V^{2+} (%)	V^{3+} (%)	V^{4+} (%)	V^{5+} (%)	VO_x composition
1	$Ar_{19}O_1^H$	0	0	74	26	$VO_{2.13}$
2	$Ar_{18.5}O_{1.5}^H$	0	0	35.9	64.1	$VO_{2.32}$
3	$Ar_{18}O_2^H$	0	0	26.8	73.2	$VO_{2.37}$
4	$Ar_{16}O_4^H$	0	0	24.7	75.3	$VO_{2.38}$
5	$Ar_{19}O_1^L$	13	80.2	6.8	0	$VO_{1.47}$
6	$Ar_{18.5}O_{1.5}^L$	0	0	51.7	48.3	$VO_{2.24}$
7	$Ar_{18}O_2^L$	0	0	49.5	50.5	$VO_{2.25}$
8	$Ar_{16}O_4^L$	0	0	47.1	52.9	$VO_{2.26}$

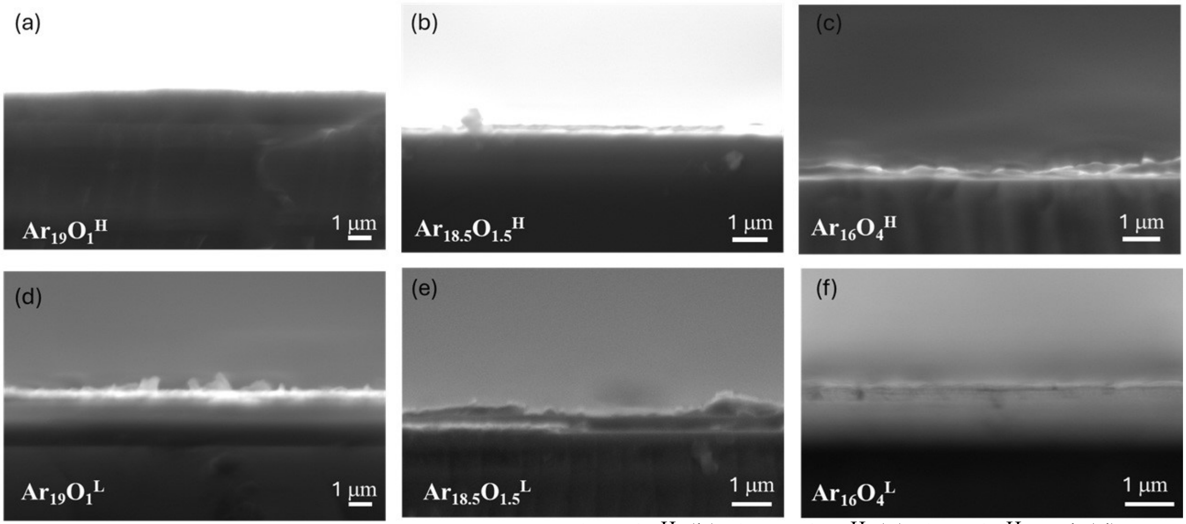

Figure S4: Cross sectional SEM micrograph of (a) $Ar_{19}O_1{}^H$ (b) $Ar_{18.5}O_{1.5}{}^H$ (c) $Ar_{16}O_4{}^H$ and (d) $Ar_{19}O_1{}^L$ (e) $Ar_{18.5}O_{1.5}{}^L$ and (f) $Ar_{16}O_4{}^L$.

Table S3: Thickness of all samples deposited at 550 ^oC and 250 ^oC.

Sample	Thickness (nm) *Error (±5 nm)*
$Ar_{19}O_1{}^H$	330
$Ar_{18.5}O_{1.5}{}^H$	285
$Ar_{18}O_2{}^H$	250
$Ar_{16}O_4{}^H$	240
$Ar_{19}O_1{}^L$	190
$Ar_{18.5}O_{1.5}{}^L$	173
$Ar_{18}O_2{}^L$	150
$Ar_{16}O_4{}^L$	158

Sample	Weight %		Atomic %	
	V	O	V	O
$Ar_{19}O_1^H$	45.8	54.2	21	79
$Ar_{18.5}O_{1.5}^H$	43.6	56.4	19.5	80.5
$Ar_{18}O_2^H$	41.8	58.2	18.1	81.9
$Ar_{16}O_4^H$	33.8	66.2	13.8	86.2
$Ar_{19}O_1^L$	25.7	74.3	9.8	90.2
$Ar_{18.5}O_{1.5}^L$	24.2	75.8	9.1	90.9
$Ar_{18}O_2^L$	23	77	9.3	90.7
$Ar_{16}O_4^L$	20.5	79.5	7.5	92.5

Table S4: EDX analysis of samples.

Table S5: TCR and sheet resistivity of films

Sample	Sheet Resistivity (Ω/sq)
$Ar_{19}O_1^H$	6.7
$Ar_{18.5}O_{1.5}^H$	4.5
$Ar_{18}O_2^H$	1.1
$Ar_{16}O_4^H$	9.5
$Ar_{19}O_1^L$	90.6
$Ar_{18.5}O_{1.5}^L$	81.5
$Ar_{18}O_2^L$	31.7
$Ar_{16}O_4^L$	55.3