## **Supporting Information**

## Improving Alkene Selectivity of Nanocarbon Catalyzed Oxidative Dehydrogenation of *n*-Butane by Refinement of Oxygen Species

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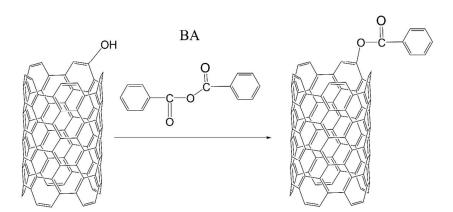
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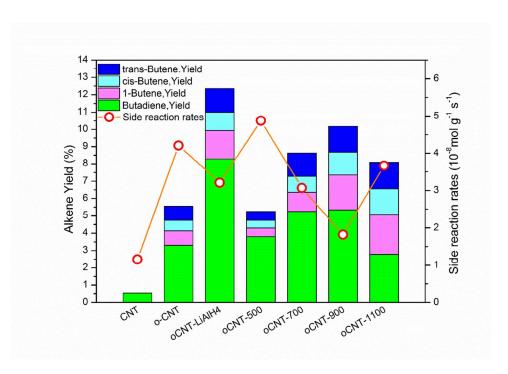
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**Table S1.** O/C weight content ratio of o-CNT, oCNT-LiAlH<sub>4</sub> and o-CNT under different annealing temperatures before and after ODH of butane.

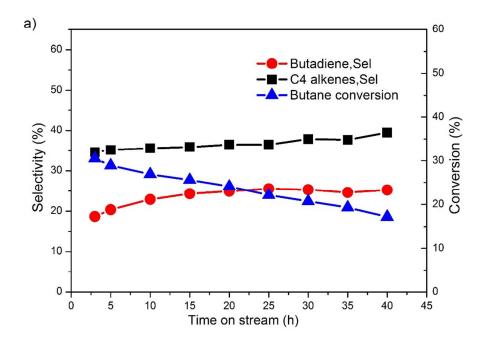
Samples	O/C weight ratio	
	Before ODH	After ODH
o-CNT	0.169	0.036
oCNT-LiAlH <sub>4</sub>	0.060	0.054
oCNT-500	0.034	0.031
oCNT-700	0.009	0.032
oCNT-900	0.004	0.017
oCNT-1100	0.003	0.020

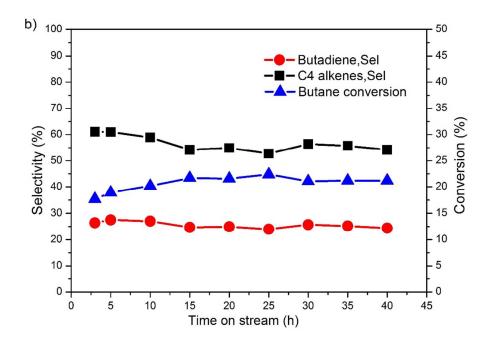


**Scheme S1.** Selective deactivation of phenol groups on CNTs by benzoic anhydride (BA).

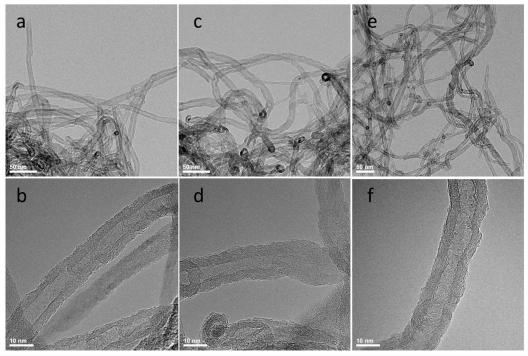


**Figure S1.** C<sub>4</sub> alkene yield and side-reaction rates of pristine CNTs, o-CNT, oCNT-LiAlH<sub>4</sub>, oCNT-500, oCNT-700, oCNT-900 and oCNT-1100. The ODH reaction conditions: 723 K, 1 atm, O<sub>2</sub>/butane=2.





**Figure S2.** Stability of a) oCNT-LiAlH<sub>4</sub> and b) oCNT-900 for ODH of butane over 40 hours. There is 55 % catalyst weight loss of catalyst of oCNT-LiAlH<sub>4</sub> after 40 hours while the catalytic performance of oCNT-900 is highly stable. The ODH reaction conditions: 723 K, 1 atm, O<sub>2</sub>/butane=2.



**Figure S3.** TEM images of o-CNT ( a,b ), oCNT-LiAlH<sub>4</sub> ( c,d ) and oCNT-900 ( e,f ).

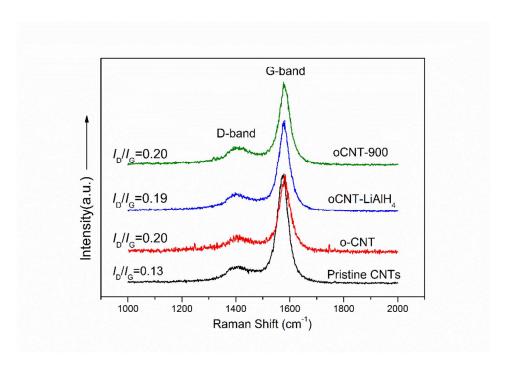
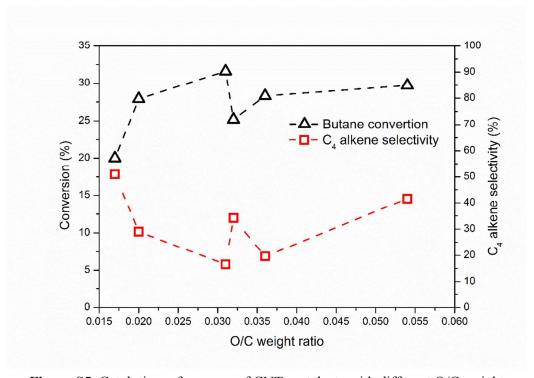
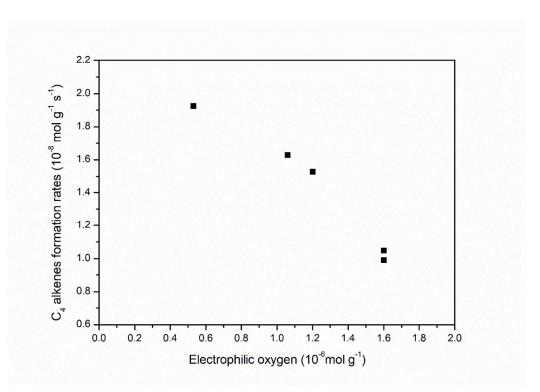


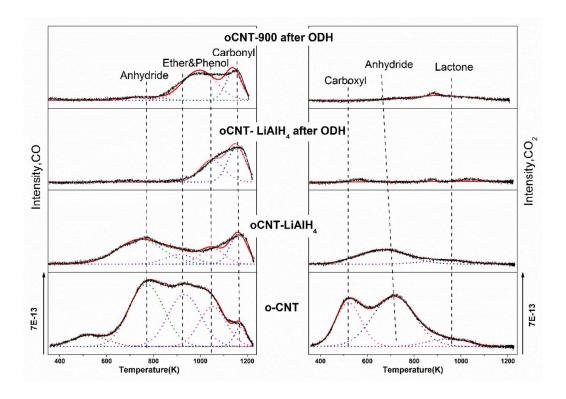
Figure S4. Raman spectra of pristine CNTs, o-CNT, oCNT-LiAlH<sub>4</sub> and oCNT-900.



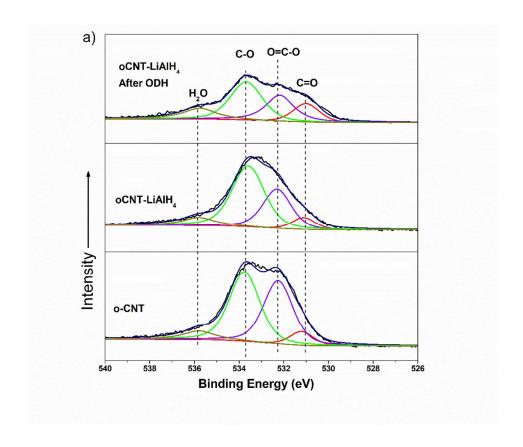
**Figure S5.** Catalytic performance of CNTs catalysts with different O/C weight content ratio. The samples involved: o-CNT, oCNT-LiAlH<sub>4</sub>, oCNT-500, oCNT-700, oCNT-900 and oCNT-1100.

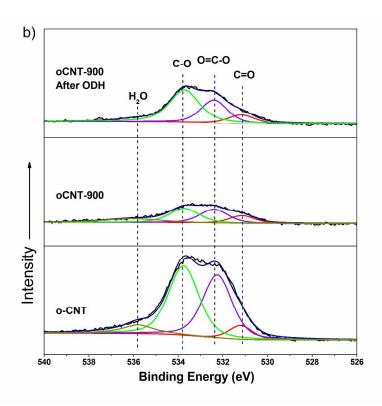


**Figure S6.** C<sub>4</sub> alkenes formation rates on CNTs samples with different amount of electrophilic oxygen after ODH of n-butane (the samples include o-CNT, oCNT-500, oCNT-700, oCNT-900 and oCNT-1100). The ODH reaction conditions: 723 K, 1 atm,  $O_2$ /butane=2.

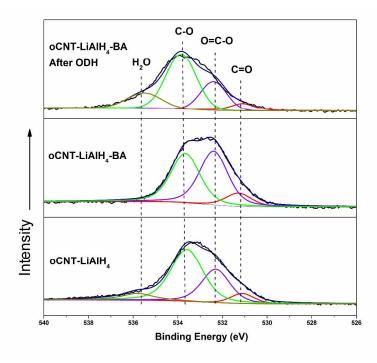


**Figure S7.** Deconvolution of TPD profiles for CO ( left ) and CO<sub>2</sub> ( right ) desorption of the following samples: o-CNT, oCNT-LiAlH<sub>4</sub>, oCNT-LiAlH<sub>4</sub> and oCNT-900 after ODH of butane.

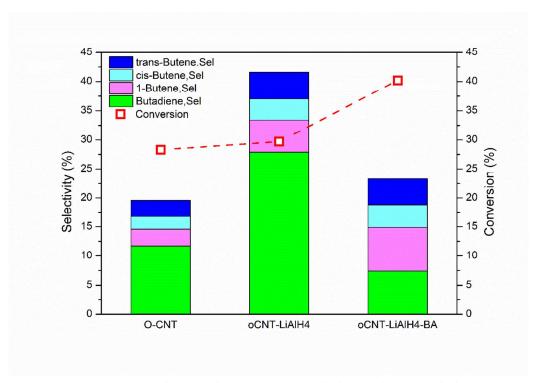




**Figure S8.** Deconvolution of O1s XPS spectra of a) o-CNT and oCNT-LiAlH<sub>4</sub> before and after ODH b) o-CNT and oCNT-900 before and after ODH.



**Figure S9.** Deconvolution of O1s XPS spectra of oCNT-LiAlH<sub>4</sub> and oCNT-LiAlH<sub>4</sub>-BA.



**Figure S10.** ODH activities of o-CNT, oCNT-LiAlH<sub>4</sub> and oCNT-LiAlH<sub>4</sub>-BA.