

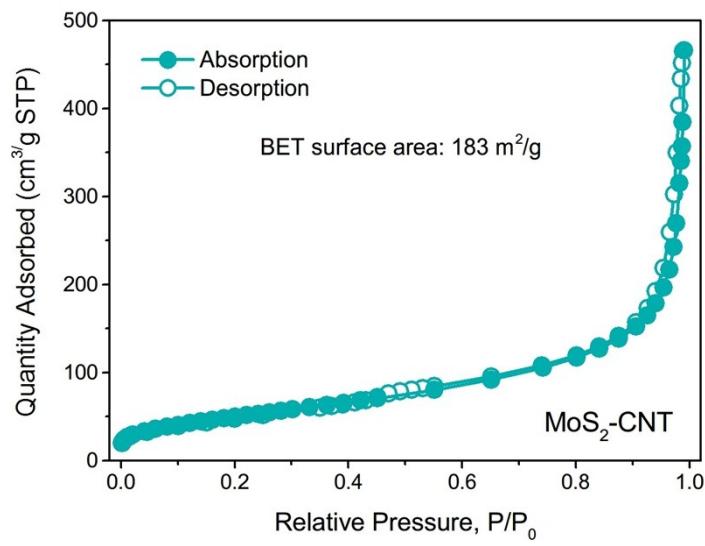
**Supporting information**

**Strongly Coupled MoS<sub>2</sub> Nanoflake-Carbon Nanotube Nanocomposite as an Excellent Electrocatalyst for Hydrogen Evolution Reaction**

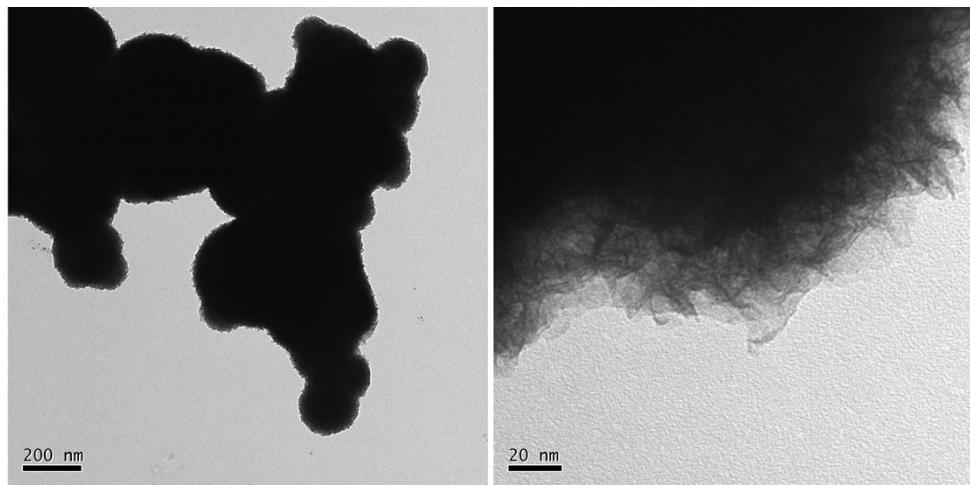
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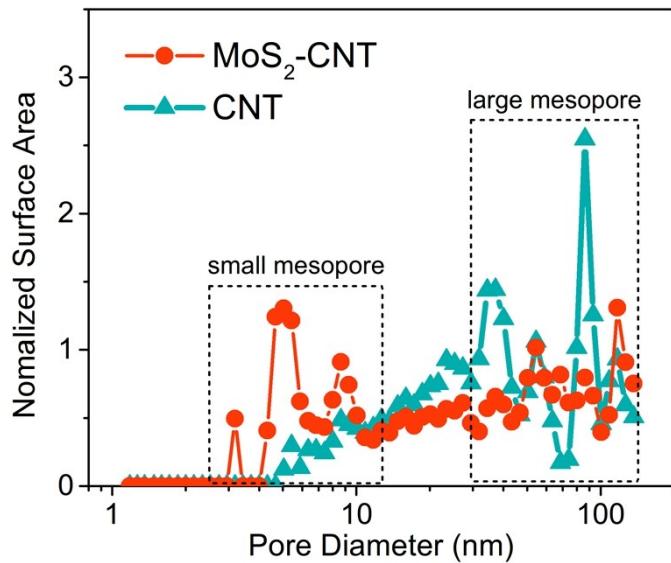
E-mail: weipeng\_liu@scau.edu.cn; liuyingju@hotmail.com



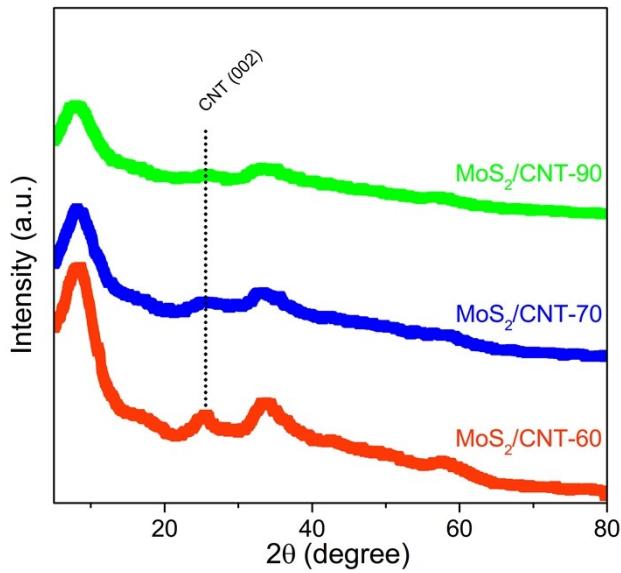
**Figure S1.** N<sub>2</sub> absorption-desorption isotherm of strongly coupled MoS<sub>2</sub>-CNT nanocomposite.



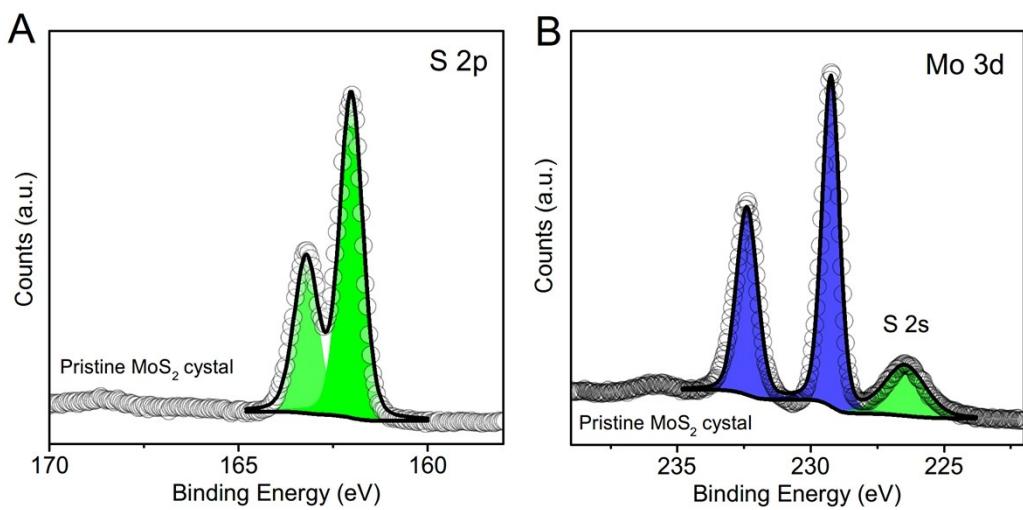
**Figure S2.** MoS<sub>2</sub> aggregates synthesized without CNT.



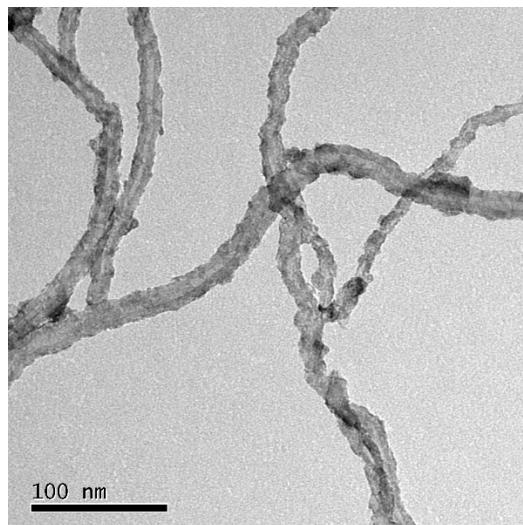
**Figure S3.** Absorption pore size distributions using DFT model of CNT and strongly coupled MoS<sub>2</sub>-CNT.



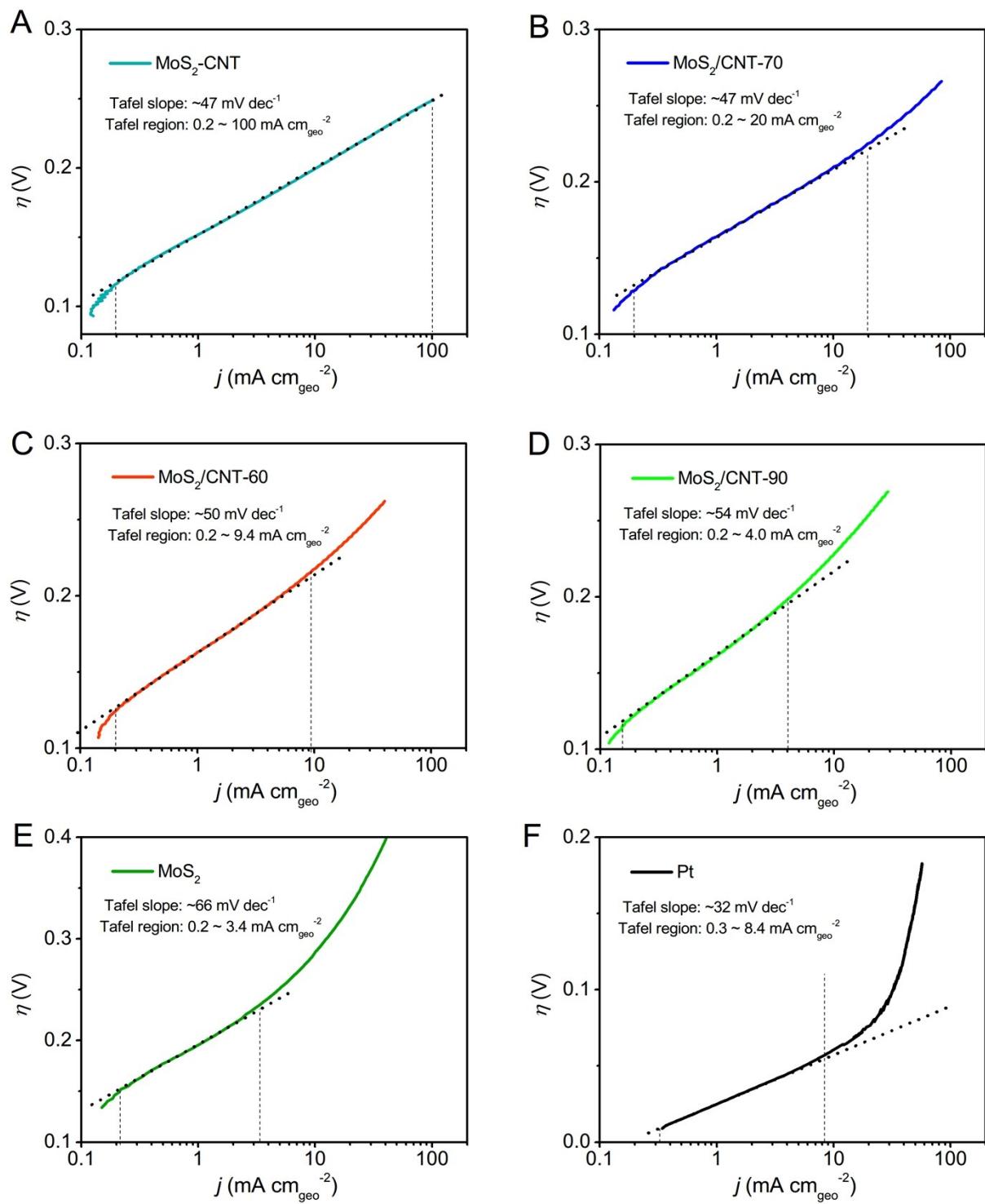
**Figure S4.** XRD patterns of MoS<sub>2</sub>/CNT-60, MoS<sub>2</sub>/CNT-70 and MoS<sub>2</sub>/CNT-90.



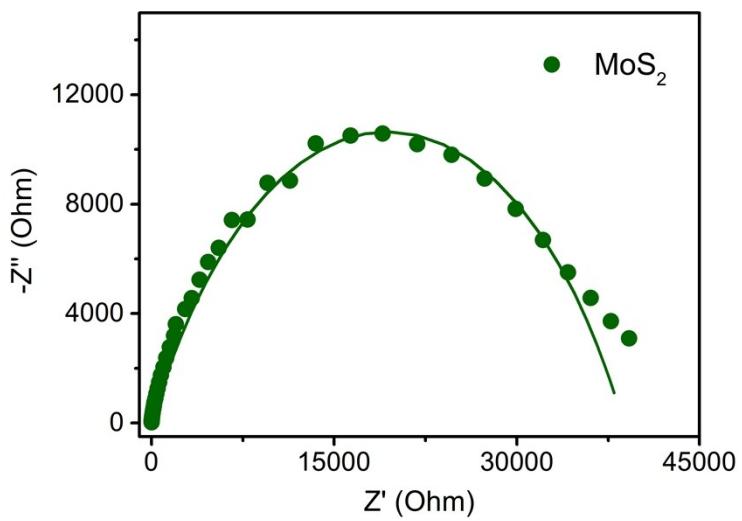
**Figure S5.** Core-level XPS spectra of pristine MoS<sub>2</sub> crystal (Aladdin, 99.5% metal basis) in the region of (A) S 2p and (B) Mo 3d.



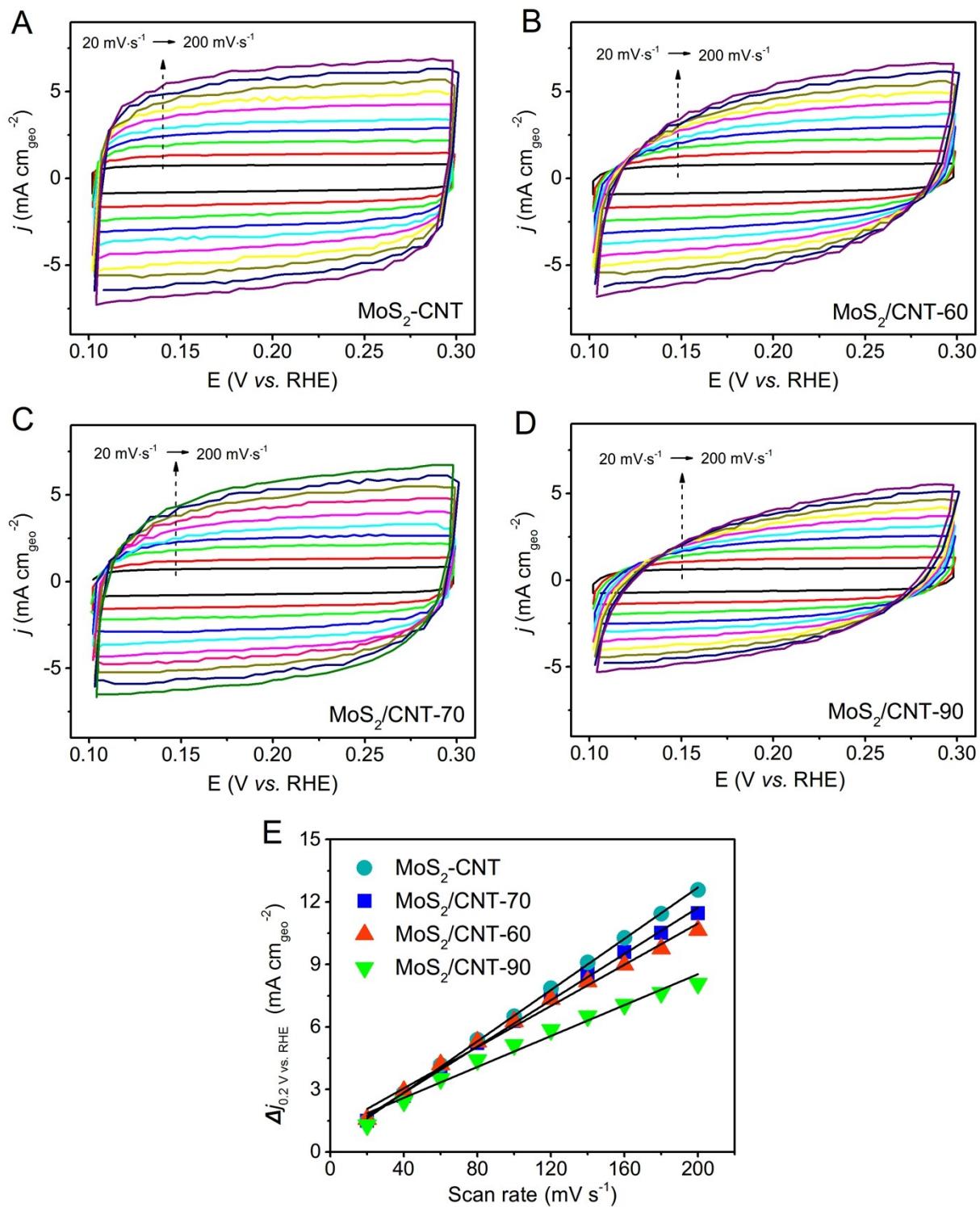
**Figure S6.** TEM image of MoO<sub>x</sub>-CNT synthesized without thiourea.



**Figure S7.** Individual Tafel plots of as-prepared samples (MoS<sub>2</sub>-CNT, MoS<sub>2</sub>/CNT-60, MoS<sub>2</sub>/CNT-70, MoS<sub>2</sub>/CNT-90 and MoS<sub>2</sub>), pristine MoS<sub>2</sub> crystal and standard Pt wire, and their measurements for Tafel slope and the length of Tafel region.



**Figure S8.** Nyquist plots of  $\text{MoS}_2$  aggregates at overpotential of 250 mV.



**Figure S9.** Cyclic voltammogram curves of (A)  $\text{MoS}_2$ -CNT, (B)  $\text{MoS}_2/\text{CNT}-60$ , (C)  $\text{MoS}_2/\text{CNT}-70$  and (D)  $\text{MoS}_2/\text{CNT}-90$  in the region of 0.1-0.3 V vs. RHE, and their differences in current density variation ( $\Delta j = j_a - j_c$ ) at potential of 0.2 V vs. RHE plotted against scan rate from 20 to 200  $\text{mV s}^{-1}$ .

**Table S1.** Comparison of HER performance of strongly coupled MoS<sub>2</sub>-CNT nanocomposite with other MoS<sub>2</sub>/nanocarbons electrocatalysts.

Catalyst	Onset η ( $\eta_0$ , mV)	Current density ( $j$ , mA cm <sup>-2</sup> )	Corresponding η ( $\eta_j$ , mV)	Electrochemical double layered capacitance ( $C_{dl}$ , mF cm <sup>-2</sup> )	R <sub>CT</sub> (Ω)	Ref.
<b>MoS<sub>2</sub>@OMC</b>	120	-10	178~192	N.A.	~900 @η=100 mV	[1]
<b>MoS<sub>2</sub>/rGO hydrogel</b>	125	-12	200	29.6	N.A.	[2]
<b>MoS<sub>2</sub>/NCNFs</b>	N.A.	-65.6	200	22.7	N.A.	[3]
<b>MoS<sub>2</sub> NPs/RGO</b>	~100	N.A.	N.A.	N.A.	~250 @η=120 mV	[4]
<b>MoS<sub>2</sub> nanoparticles on mesoporous graphene foams</b>	100	-100	200	N.A.	33 @η=150 mV	[5]
<b>MoS<sub>2</sub> NSs/RGO</b>	~140	-23	200	N.A.	155 @η=170 mV	[6]
<b>MoS<sub>2</sub> ⊥ RGO</b>	N.A.	-10	172	6.045	N.A.	[7]
<b>MoS<sub>x</sub> on crumpled graphene balls</b>	130	-220	300	N.A.	N.A.	[8]
<b>MoS<sub>x</sub>/N-doped CNT</b>	~75	-10	~110	N.A.	~150 @η=200 mV	[9]
<b>MoS<sub>2</sub>/VGNS</b>	160	N.A.	N.A.	7.96	39.2 @open circuit	[10]
<b>MoS<sub>2</sub>/CA</b>	140	-9.68	200	N.A.	N.A.	[11]
pBC-N/MoS <sub>2</sub>	108	-8.7	200	16.5	N.A.	[12]
<b>C/MoS<sub>2</sub>@G</b>	165	-100	~360	N.A.	13.2 @η=300 mV	[13]
<b>Strongly coupled MoS<sub>2</sub>-CNT</b>	~100	-850	290	31	12 @η=250 mV	This work
		-15	208			

## References

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