Supplementary materials

High-Entropy Alloy Nanoparticles combining SiC coating Synergistically Boost Electromagnetic Shielding Performance of Carbon Nanotube Sponge

Ying Zhang, Chengqing Tang, Yaoqieyu Song, Sheng Zhang, Zhi Hong Hang,* Xiaohua Zhang,* Yitan Li,* and Zhaohui Yang*

 School of Physical Science and Technology, Soochow University, Suzhou, 215006, P.R. China.
National Engineering Research Center for Colloidal Materials, School of Chemistry and Chemical Engineering, Shandong University, Jinan, Shandong, 250100, P. R. China.

3. Key Laboratory of Colloid and Interface Chemistry of the Ministry of Education, Shandong University, Jinan, Shandong, 250100, P. R. China

 Center for Soft Condensed Matter Physics and Interdisciplinary Research, Soochow University, Suzhou, 215006, P. R. China.

5. Institute for Advanced Study, Collaborative Innovation Center of Suzhou Nano Science and Technology, Soochow University, Suzhou, 215006, P. R. China

Corresponding Authors

E-mail address : <u>zhhang@suda.edu.cn</u>(Z.H.Hang)

E-mail address : <u>zhangxiaohua@suda.edu.cn</u> (X.H.Zhang)

E-mail address : <u>yitanli@sdu.edu.cn</u> (Y.T.Li)

E-mail address : <u>yangzhaohui@suda.edu.cn</u> (Z.H.Yang)

Additional figures and tables



Figure S1. Low density sponge loaded HEA

The carbon nanotube sponges with low density ($< 0.15 \text{ g/cm}^3$) are prone to be brittle and hard after loading with HEA.



Figure S2. Image of CNTS@HEA

The black surface of CNTS turns into a metallic luster indicating the deposition of HEAs.



Figure S3. Image of CNTS@HEA@SiC

After the deposition of SiC layer, the composite appears to light blue due to the interference of the visible light on the thin SiC layer.



Figure S4. Size distributions of HEAs

We selected 20 HEA nanoparticles and made the histogram of size distributions. The diameters of HEA are distributed between 10-40 nm, which is consistent with the SEM observation.



Figure S5.EDS of CNTS@HEA.



Figure S6.EDS of CNTS@HEA@SiC

The EDS surface elemental analysis, specific elemental distribution and energy maps of CNTS@HEA and CNTS@HEA@SiC was shown in the fig. S5,S6. It can be seen that the elements of Fe, Co, Ni, Cu, Al and Si are distributed evenly across the CNTS.



The characteristic peaks of each metal, C1s, O1s and Si2p in CNTS@HEA can be clearly distinguished in the XPS full spectrum.



In Fig S8, it can be seen that with the addition of SiC, the R/A values of the materials all increase to some extent. It indicates that the formation of Schottky junction promotes the reflection effect of electromagnetic wave and enhances the loss of electromagnetic waves.



Figure S9. Size distributions of HEAs annealed at high temperatures

We selected 20 HEA nanoparticles and made the histogram of size distributions. The diameters of HEA are smaller at high annealing temperatures compared with **Fig.S4**.



Figure S10.XRD of CNTS@Isomolar annealed at 500 800 and 1000°C

In XRD analysis, their crystal lattice was changed in high temperature. These may account for some reduction in the shielding performance of the samples at high temperatures, but in general it remains at a high level.



Figure S11.XRD of CNTS@Isomolar and CNTS@Isomolar@SiC acidified by nitric acid separately

In XRD analysis, the HEA peaks of CNTS@Isomolar nearly disappear, approximating the pure carbon nanotube sponges. The HEA peaks of CNTS@Isomolar@SiC sample still existed, confirming the excellent protection of the SiC shell.

Sample number	Sample quality m ₀ (g)	Constant volume V ₀ (mL)	Test Elements	Test Solution Element ConcentrationC _o (ug/L)	dilution factor f	Elemental concentration of the original solution of the digestion solution	Sample Elemental Content C _x (ug/kg)	Sample Elemental Content W (%)
1	0.0523	25	Fe	198.83	1000	198834.02	95044944	9.50%
1	0.0523	25	Fe	198.39	1000	198392.32	94833804	9.48%
2	0.0573	25	Fe	208.93	1000	208933.06	91157532	9.12%
2	0.0573	25	Fe	208.82	1000	208820.19	91108287	9.11%
1	0.0523	25	Co	70.71	1000	70709.73	33800061	3.38%
1	0.0523	25	Co	70.67	1000	70667.14	33779703	3.38%
2	0.0573	25	Co	143.40	1000	143395.99	62563694	6.26%
2	0.0573	25	Co	143.19	1000	143188.71	62473259	6.25%
1	0.0523	25	Ni	75.24	1000	75242.83	35966935	3.60%
1	0.0523	25	Ni	75.86	1000	75856.25	36260156	3.63%
2	0.0573	25	Ni	93.82	1000	93822.05	40934579	4.09%
2	0.0573	25	Ni	93.58	1000	93583.32	40830417	4.08%
1	0.0523	25	Cu	80.17	1000	80172.26	38323258	3.83%
1	0.0523	25	Cu	80.26	1000	80263.56	38366901	3.84%
2	0.0573	25	Cu	46.72	1000	46724.32	20385830	2.04%
2	0.0573	25	Cu	46.59	1000	46591.10	20327703	2.03%
1	0.0523	25	Al	329.74	100	32974.42	15762152	1.58%
1	0.0523	25	Al	329.89	100	32989.14	15769188	1.58%
2	0.0573	25	Al	175.36	100	17536.05	7650983	0.77%
2	0.0573	25	Al	175.38	100	17537.74	7651721	0.77%

Table S1. Inductively coupled plasma-mass spectrometer (ICP-MS) summarized data

We prepared 50mg of Sample CNTS@Isomolar and Sample CNTS@Co₂, denoted by number 1 and 2, respectively. We can get the elemental content of the samples. The Co content in CNTS@Co₂ sample is approximated to be twice as much as CNTS@Isomolar sample, and the content of Cu and Al in CNTS@Co₂ sample is about 1/2 as that of CNTS@Isomolar sample, which is in line with the expectation. The content of each metal is consistent with the feeding amount, indicating the successful synthesis of HEAs.