

Supplementary information for :

## High efficient large-scale preparation and electromagnetic property control of silica-NiFeP double shell composite hollow particles

Bin Liao,<sup>a,b</sup> Zhenguo An,<sup>\*a</sup> Jingjie Zhang<sup>\*,a</sup>

<sup>a</sup>Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190 China.

<sup>b</sup>University of Chinese Academy of Sciences, Beijing 100049 , P. R. China.

Agents and factors (alkaline solution)	S1				S2				S3			
NiSO <sub>4</sub> ·6H <sub>2</sub> O	66.6				44.7				33.5			
(NH <sub>4</sub> ) <sub>2</sub> Fe(SO <sub>4</sub> ) <sub>2</sub> ·6H <sub>2</sub> O	0				33.3				50			
NaH <sub>2</sub> PO <sub>2</sub> ·H <sub>2</sub> O	25				25				25			
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	10				10				10			
KNaC <sub>4</sub> H <sub>4</sub> O <sub>6</sub> ·4H <sub>2</sub> O	75				75				75			
NH <sub>2</sub> CH <sub>2</sub> COOH	10				10				10			
PEG	10				10				10			
pH value	9				9				9			
temperature/°C	70				70				70			
Mole ratio(Fe <sup>2+</sup> /(Fe <sup>2+</sup> +Ni <sup>2+</sup> ))	0				1:3				1:2			
Sample Names	S11	S12	S13	S14	S21	S22	S23	S24	S31	S32	S33	S34
Volume of solution(mL)	400	300	200	100	400	300	200	100	400	300	200	100
Mass of seeds(g)	4g	3/4S11	2/3S12	1/2S13	4g	3/4S21	2/3S22	1/2S23	4g	3/4S31	2/3S32	1/2S33

**Table S1** Detailed chemical agent concentrations (g/L) and reaction conditions for different samples.

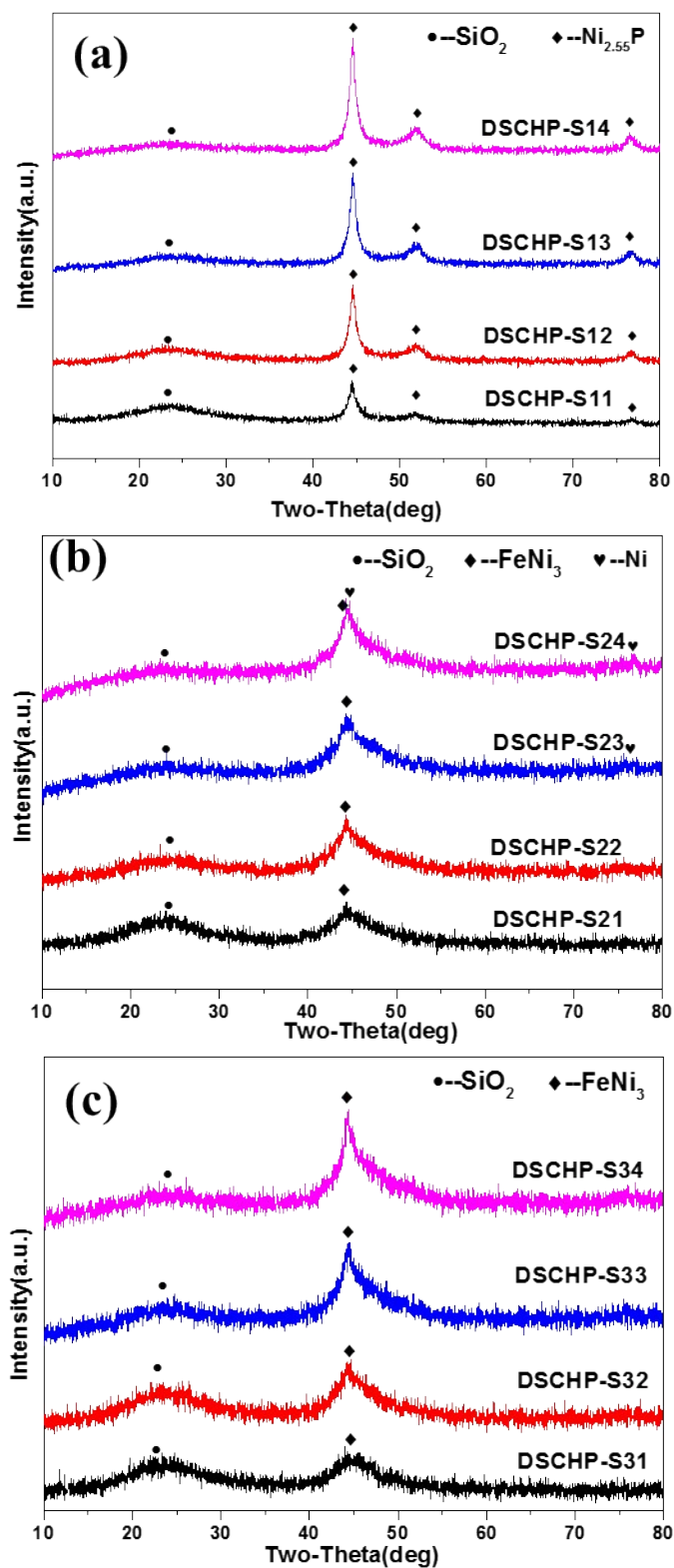
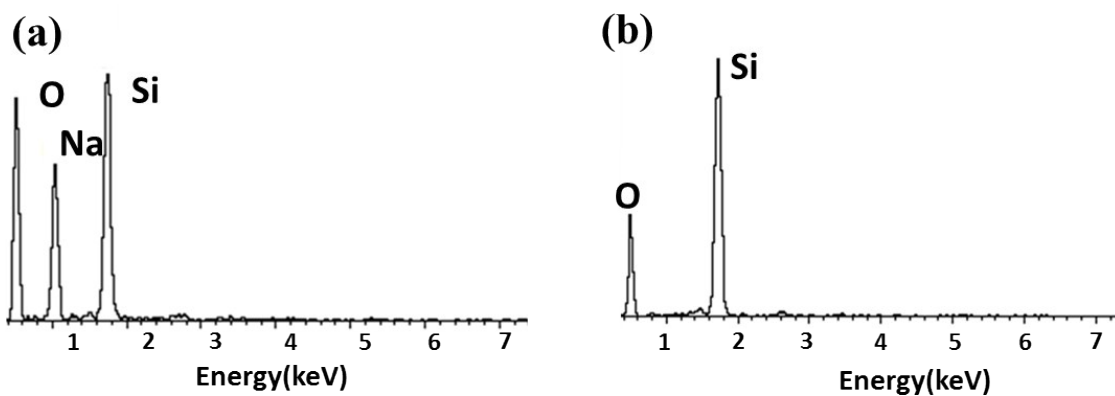
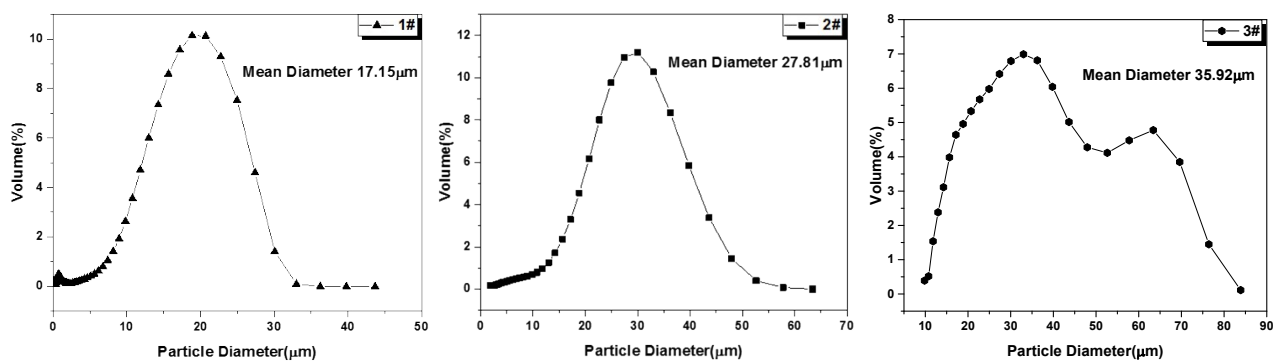


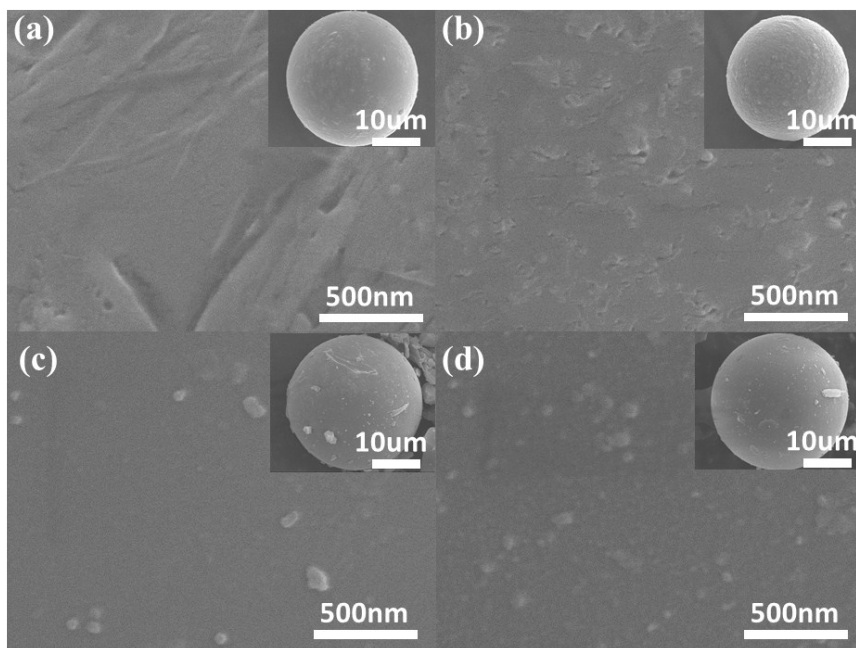
Fig. S1 XRD patterns of (a) DSCHP-S1, (b) DSCHP-S2 and (c) DSCHP-S3 with different plating times



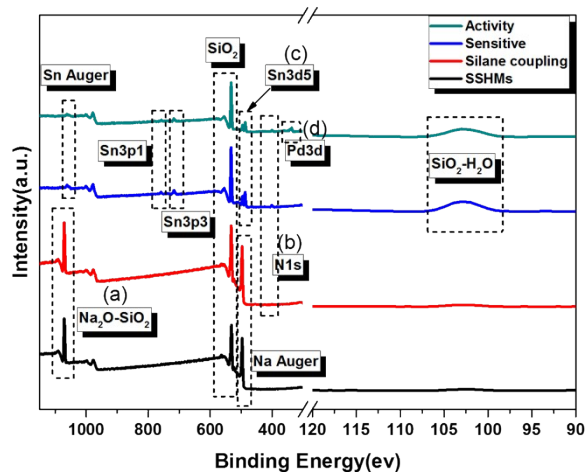
**Fig. S2** EDX spectra of the obtained hollow particles with treatment (a) silane coupling and (b) sensitization



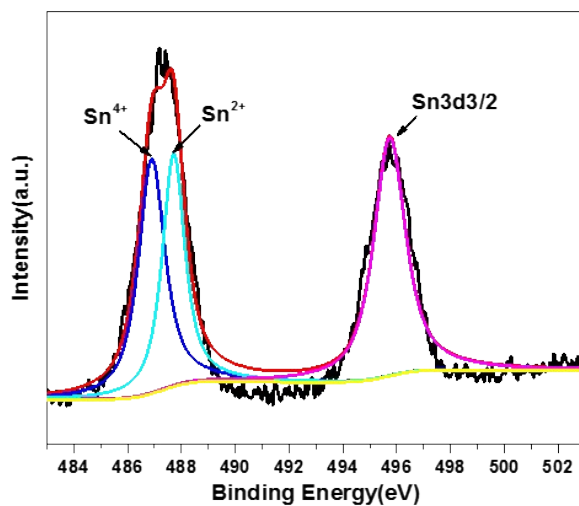
**Fig. S3** Particle size distribution of obtained SSHPs with different parameter of spray drying.



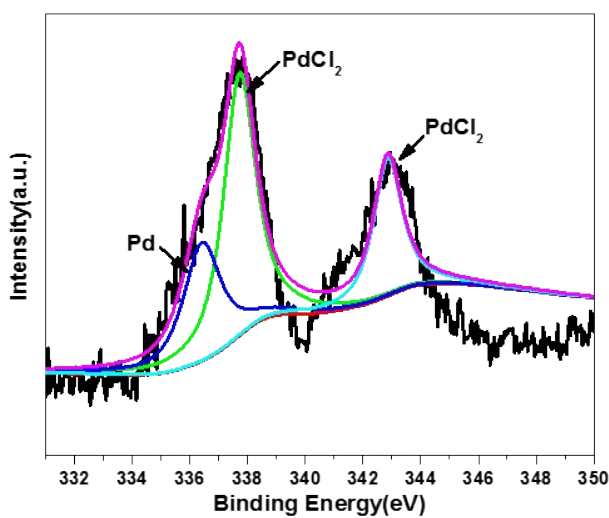
**Fig. S4** SEM images of the hollow particles at different stage of pretreatments. (a) the pristine SSHPs, SSHPs obtained after silane coupling (b), sensitization (c) and activation (d).



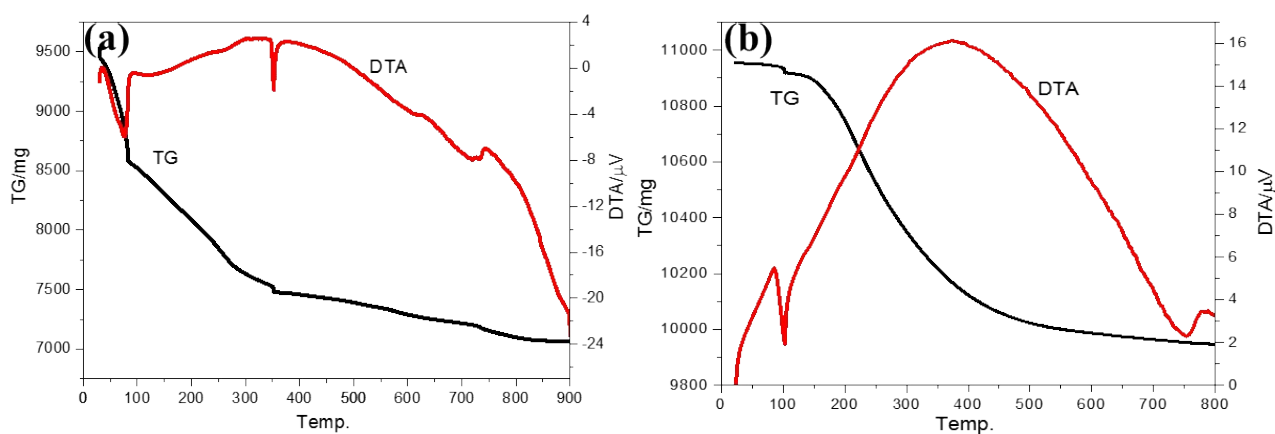
**Fig. S5** XPS spectra of the SSHPs samples at different stage of pretreatment: (a) pristine SSHPs, (b) SSHPs after silane coupling, (c) SSHPs after activation.



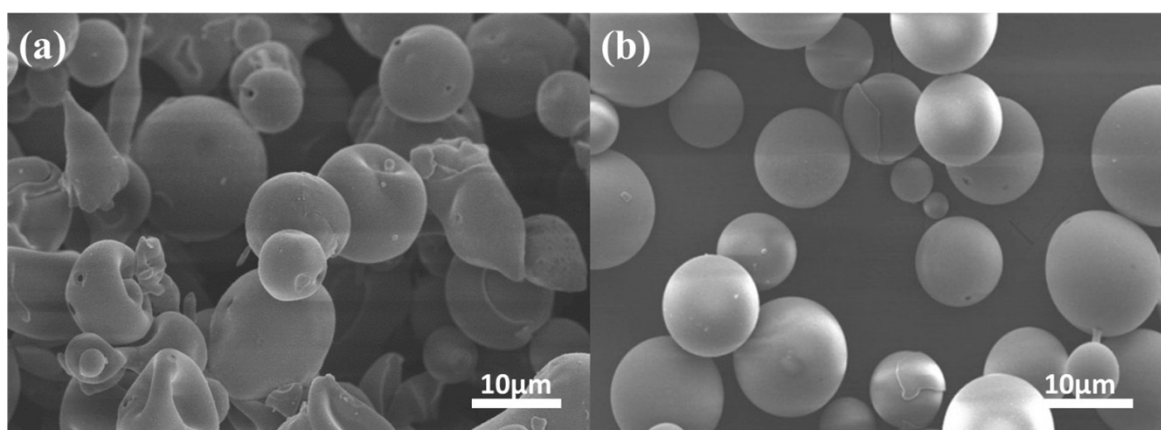
**Fig. S6** XPS spectra of Sn3d for SSHPs after activation.



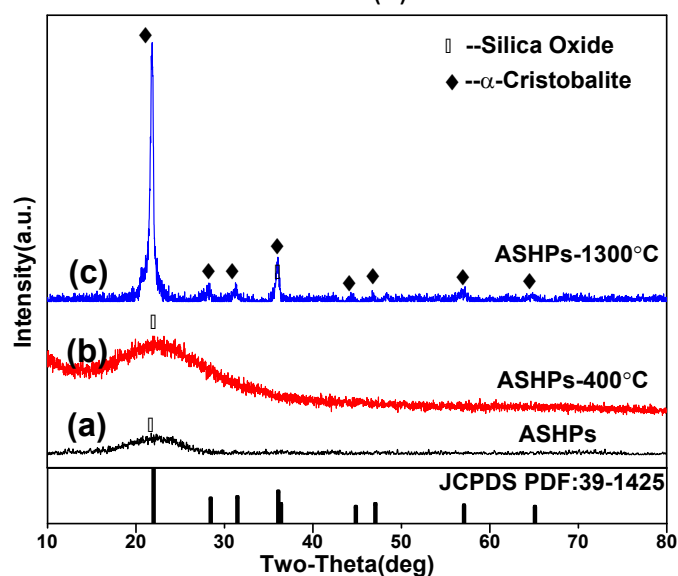
**Fig. S7** XPS spectra of Pd3d for SSHPs after activation.



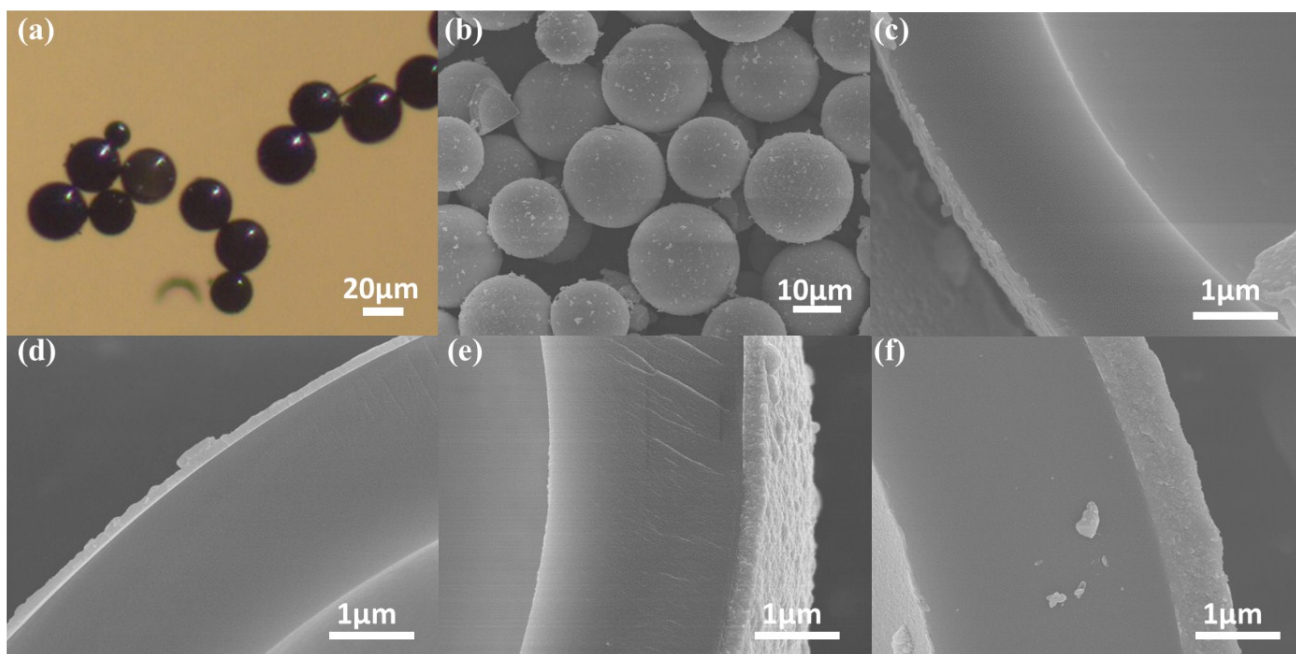
**Fig. S8** TG-DTA curves of (a) SSHPs and (b) ASHPs.



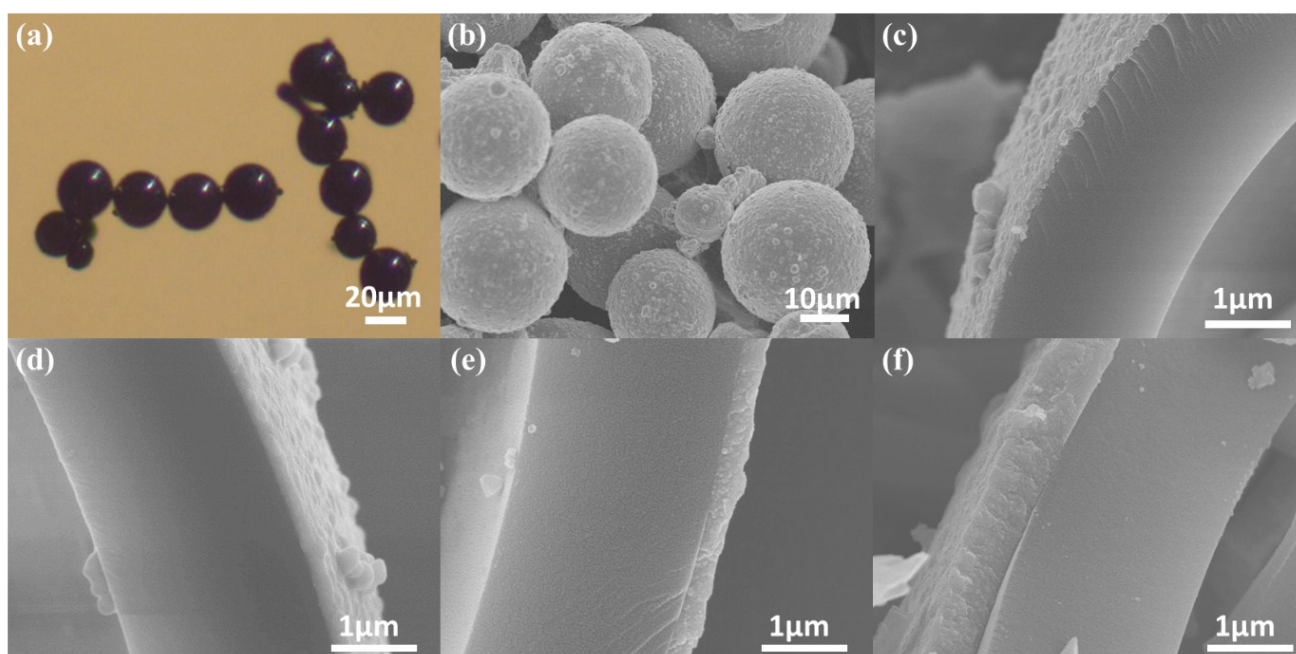
**Fig. S9** SEM images of the pristine SSHPs calcinated at 400°C (a) and the ASHPs calcinated at 1300°C (b).



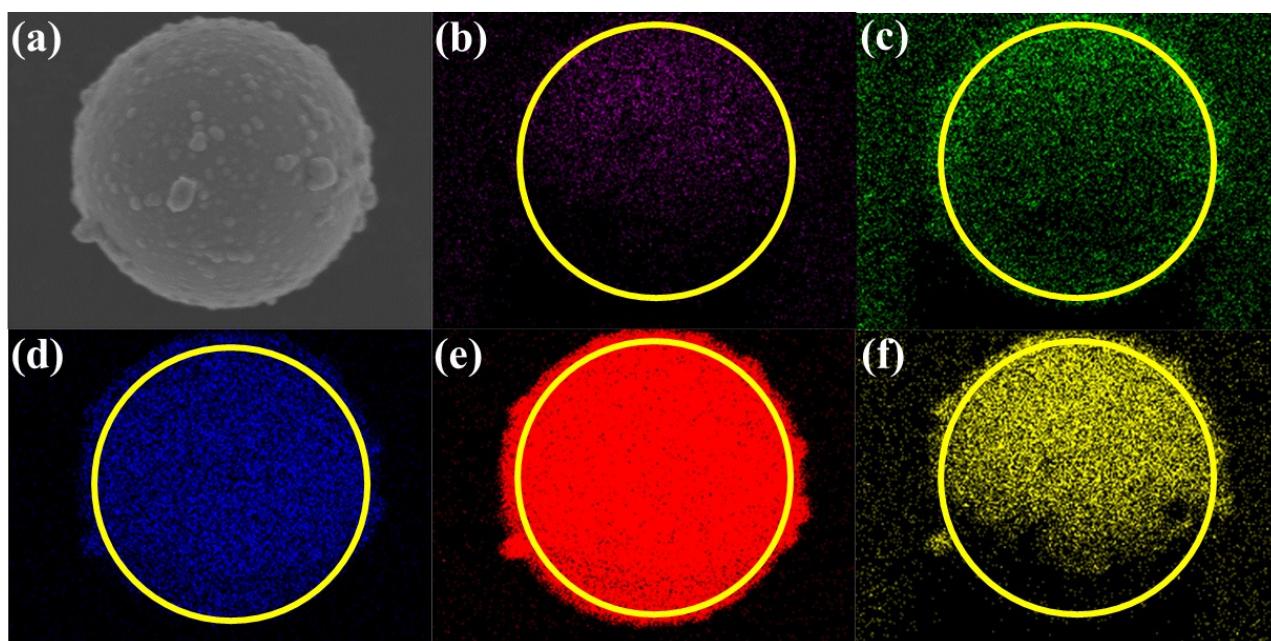
**Fig. S10** XRD patterns of the ASHPs with heat treatment: (a)ASHPs, (b)400°C, and (c)1300°C



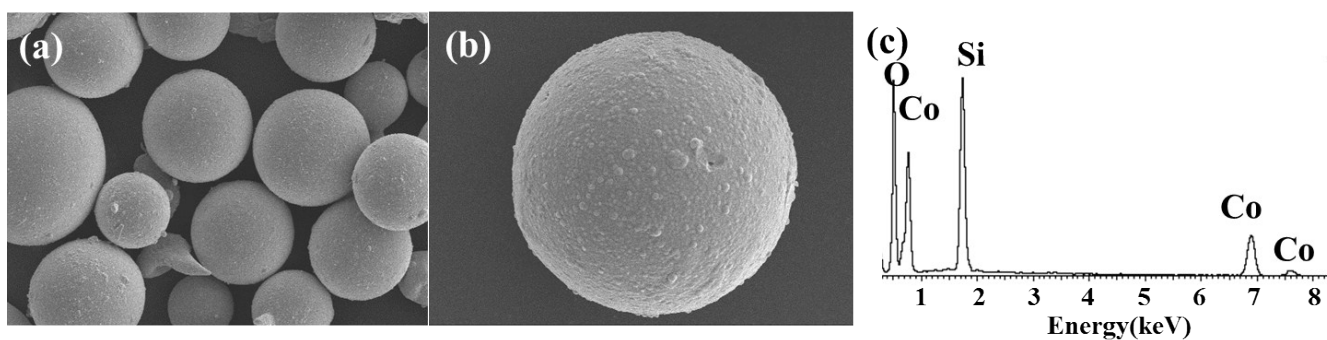
**Fig. S11** (a) Optical microscopy image of DSCHP-S11 and SEM images of (b), (c) DSCHP-S11, (d) DSCHP-S12, (e)DSCHP-S13, (f)DSCHP-S14.



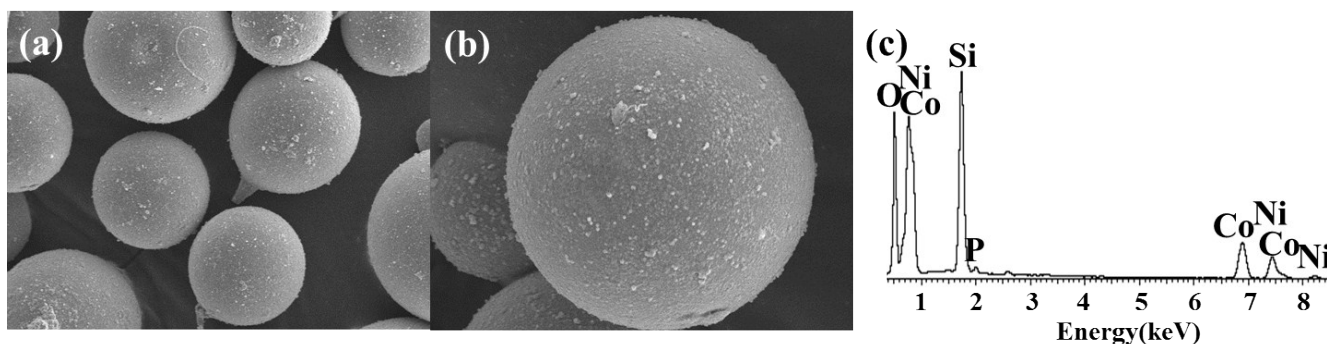
**Fig. S12** (a) Optical microscopy image of DSCHP-S31 and SEM images of (b), (c) DSCHP-S31, (d) DSCHP-S32, (e)DSCHP-S33, (f)DSCHP-S34.



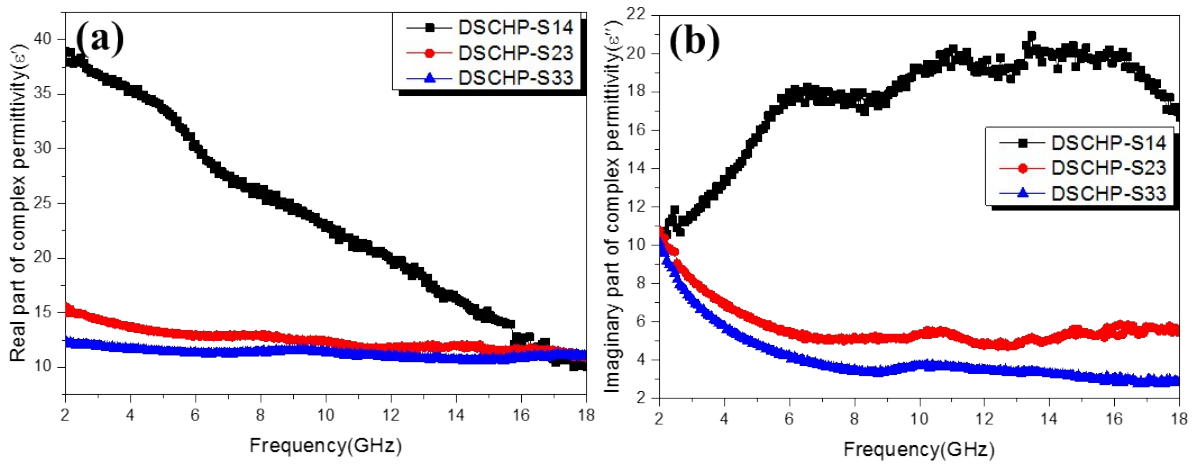
**Fig. 13** SEM (a) image and the corresponding EDX element mapping of (b) Si, (c) O, (d) Fe and (e) Ni, and (f) P taken on a single particle of the as-obtained DSCHP-S23.



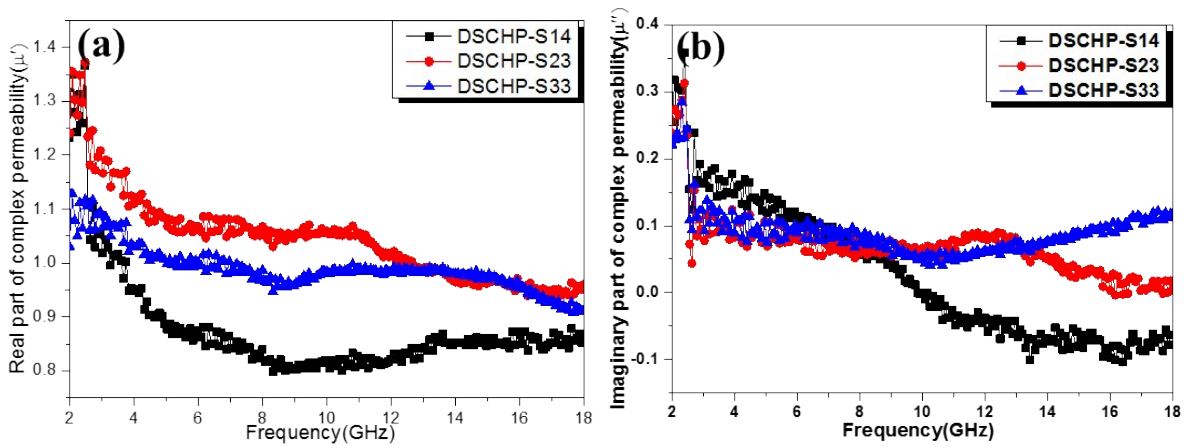
**Fig. S14** (a), (b) SEM images and (c) EDX spectra of the as-obtained  $\text{SiO}_2$ -Co composite hollow particles.



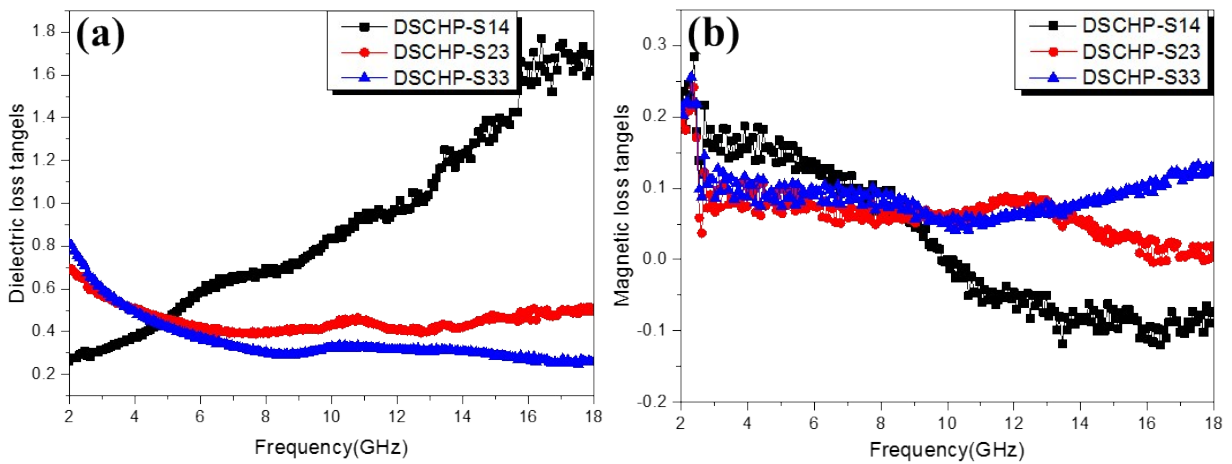
**Fig. S15** (a), (b) SEM images and (c) EDX spectra of the as-obtained  $\text{SiO}_2$ - NiCoP composite hollow particles.



**Fig. S16** The (a) real parts ( $\epsilon'$ ) and (b) imaginary parts ( $\epsilon''$ ) of DSCHP-S14 (black curve), DSCHP-S23 (red curve) and DSCHP-S33 (blue curve).

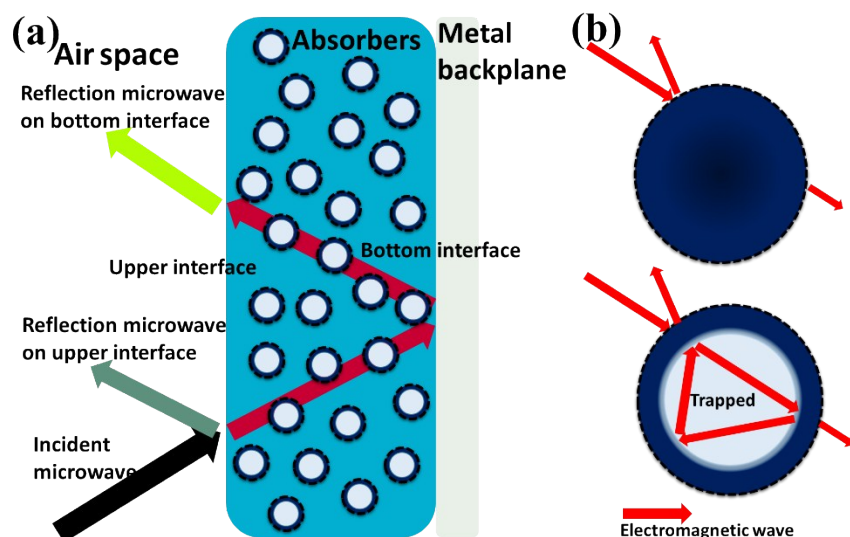


**Fig. S17** The (a) real ( $\mu'$ ) and (b) imaginary ( $\mu''$ ) parts of the permeability of DSCHP-S14 (black curve), DSCHP-S23 (red curve) and DSCHP-S33 (blue curve).



**Fig. S18** Frequency dependences of the (a) dielectric loss tangents, (b) magnetic loss tangents





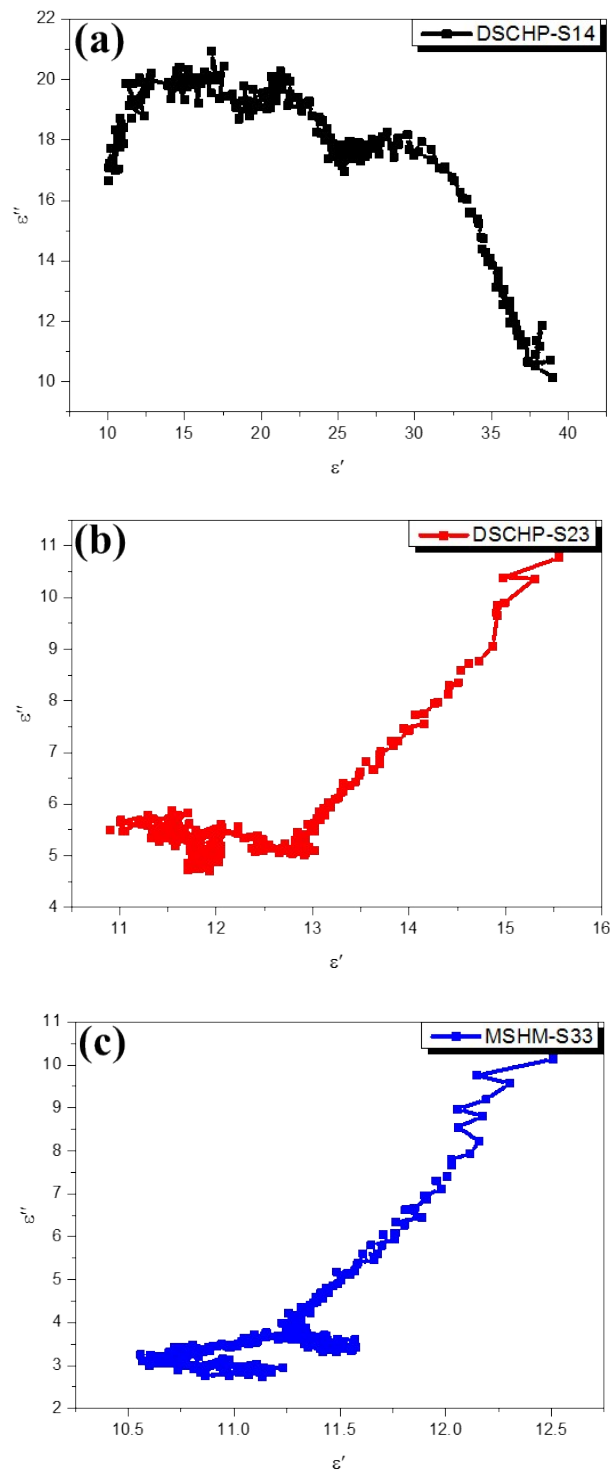
**Fig. S19** The schematic diagram of electromagnetic microwave absorption mechanism for the DCHPs. (a) electromagnetic wave incident to single-layer absorber backed by a metal backplane; (b) electromagnetic wave pathway illustration of solid structure and hollow structure.

Sample	Thickness (mm)	RL value (dB)	Bandwidth (RL<-10dB)(GHz)	Content (%)	Reference
Hollow ceramic-barium ferrites composite single shell microspheres	2.5	-10	<1	60	1
Hollow CoFe <sub>2</sub> O <sub>4</sub> /Co <sub>3</sub> Fe <sub>7</sub> microspheres	2	-41.6	3	66.7	2
CoFe <sub>2</sub> O <sub>4</sub> hollow sphere/graphene	2	-18.5	3.7	60	3
Co <sub>3</sub> Fe <sub>7</sub> /C nanoparticle	1.8	-26.8	2	50	4
FeCo/Graphene nanoparticle	2.5	-40.2	4	60	5
Hollow glass/nickel flowers microspheres	3.5	-15	0.8	30	6
NiCoP hollow spheres	2.5	-41.7	2	40	7
Silica@Iron microcubes	4.5	-54	1.5	55	8
Ni-15%Fe coated microspheres	5	-25	<1	50	9
Co-Co <sub>3</sub> O <sub>4</sub> hybrid hollow sphere	5	-18.5	<1	30	10
C@Ni-NiO solid sphere	5	-10	<1	30	11
Hollow glass/Ni thin film	3	-35	1	37.5	12
DSCHPs-S23	1.75	-16.46	3.2	30	This work
DSCHPs-S33	1.5	-45.9	4	30	This work

**Table S2.** The electromagnetic absorption properties of similar samples

## Reference

- 1 X. Cai, J. Wang, R. Yang, C. Xiong, H. Lou, *Int. J. Appl. Ceram. Technol.*, 2015, **12**, 8.
- 2 W. Li, L. Wang, G. Li, Y. Xu, *J. Magn. Magn. Mater.*, 2015, **377**, 259.
- 3 M. Fu, Q. Jiao, Y. Zhao, H. Li, *J. Mater. Chem. A*, 2014, **2**, 735.
- 4 X. D. Guo, X. J. Qiao, Q. G. Ren, X. Wan, W. C. Li, Z. G. Sun, *Appl. Phys. A*, 2015, **120**, 43.
- 5 X. Li, J. Feng, Y. Du, J. Bai, H. Fan, H. Zhang, Y. Peng, F. Li, *J. Mater. Chem. A*, 2015, **3**, 5535.
- 6 Z. An, S. Pan, J. Zhang, *J. Phys. Chem. C*, 2009, **113**, 2715.
- 7 Z. Li, Y. Deng, B. Shen, L. Liu, W. Hu, *J. Alloy. Compd.*, 2010, **491**, 406.
- 8 Z. Yang, Z. Li, L. Yu, Y. Yang, Z. Xu, *J. Mater. Chem. C*, 2014, **2**, 7583.
- 9 S. T. Kim, S. S. Kim, *J. Appl. Phys.*, 2014, **115**, 17A528.
- 10 H. Wu, Q. Wu, L. Wang, *Mater. Character.*, 2015, **103**, 1.
- 11 H. Wu, G. Wu, Q. Wu, L. Wang, *Mater. Character.*, Wang, 2014, **97**, 18.
- 12 Z. W. Liu, L. X. Phua, Y. Liu, C. K. Ong, *J. Appl. Phys.*, 2006, **100**, 093902.



**Fig. S20** Typical Cole-Cole semicircles for (a) DSCHP-S14, (b) DSCHP-S23 and (c) DSCHP-S33.