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Rational design of multiple-layer films with continuous impedance gradient variation for enhanced microwave absorption performances

Bin Quan,[a] Yu Wang,[a] Yu Chen,[a] Mingji Zhang,[b] Jun Liu,[c] Qi Jia,[c] Xiaochi Lu,[d] Jun Chen,[e] Xiaogu Huang*[a]

[a] *School of Chemistry and Materials Science, Nanjing University of Information Science & Technology, Nanjing 210044, China*

[b] *Sino-German College of Intelligent Manufacturing, Shenzhen Technology University, Shenzhen 518118, China*

[c] *College of Field Engineering, Army Engineering University of PLA, Nanjing 210094, China*

[d] *College of electronic and optical engineering & college of flexible electronics (future technology), Nanjing University of Posts and Telecommunications, Nanjing 210023, China*

[e] *Jiangsu Jewel Technology Group Co., LTD, Wuxi 214000, China*

*Corresponding authors:

Prof. Bin Quan
E-mail: binquan@nuist.edu.cn;

Prof. Xiaogu Huang
E-mail: hxg@nuist.edu.cn

Preparation of GCu samples

The schematic of preparation process is presented in Fig. S1. The 100 mg graphene oxide (GO) were dissolved in 200 mL ethanol by ultrasound. The $Cu(NO_3)_2 \cdot 3H_2O$ (1.9 g) and trimesic acid (1.7 g) were dissolved in 100 mL deionized water and 100 mL ethanol, respectively. Then trimesic acid solution and GO solution were added to $Cu(NO_3)_2$ solution in turn, and stirred for 18 hours. The GO/HKUST-1 films were prepared through vacuum filtration, and peeled from polyvinylidene fluoride filter membrane. The wet product was dried in vacuum oven for 30 min, and then the dried films were annealed under N_2 atmosphere for 3 h with a heating rate of 5°C/min to obtain the GCo films. GCu samples that were annealed at 400°C, 600°C, and 800°C were labeled as GCu400, GCu600, and GCu800, respectively.

Preparation of toroidal testing rings

For the fabrication of paraffin/film/paraffin structures, liquid wax was previously prepared with continuous base heating. The entire preparation process was carried out in the in a grinding cutter with standard toroidal shape (inner diameter of 3.04 mm, outer diameter of 7.00 mm). Firstly, a certain amount of tightly stacked multiple-layer films (the same cross section as the standard toroidal shape) were filled into the gap of grinding cutter. Then, liquid paraffin was poured into the grinding cutter, making full contact with the pre-added multiple-layer films. Finally, when the liquid paraffin was solidified, subtle treatment was conducted to obtain a standard testing sample with paraffin/film/paraffin structures. It is worth noting that, in order to maintain the mass fraction of multiple-layer films to paraffin as 1:9, the addition of multiple-layer films and liquid wax were both previously calculated. For the convenience of contrast experiment, thickness of all the testing samples were kept as 2 mm.

Table. S1 Magnetic properties of GCo samples.

Sample	M_s (emu/g)	M_r (emu/g)	H_c (Oe)
GCo400	2.2950	0.6679	378
GCo600	2.4503	0.3085	200
GCo800	34.4056	0.6072	50

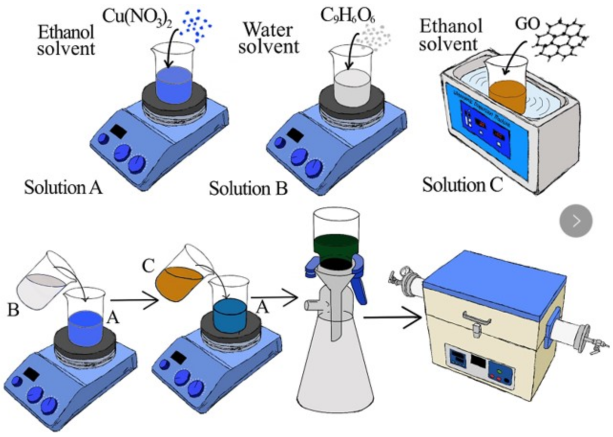

Fig. S1 Schematic illustration of the synthesis process of the GCu films.

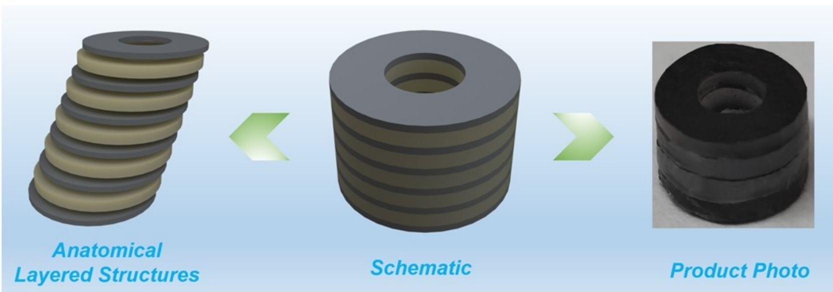

Fig. S2 Schematic of layered paraffin/film/paraffin testing sample and product photo.

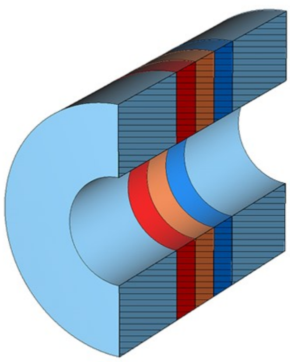

Fig. S3 Schematic diagram of three-layer model for CST simulation.

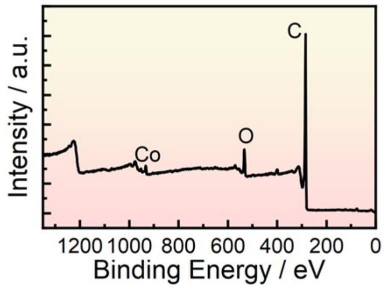

Fig. S4 XPS survey scan spectra of GCo600 films.

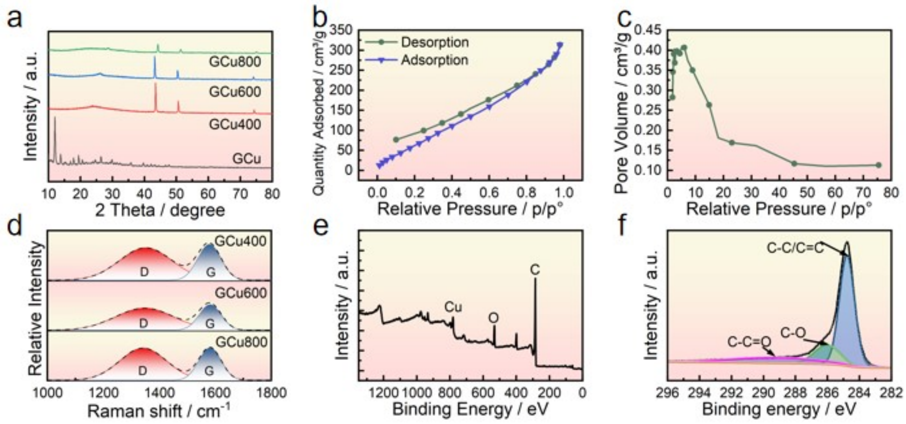

Fig. S5 (a) XRD patterns and (d) Raman spectra of GCu samples. (b) Isotherms of N_2 adsorption/desorption and (c) the pore-size distribution of GCu600 samples. (e) XPS survey scan spectra, and (e) XPS C 1s spectra of GCu600 samples.

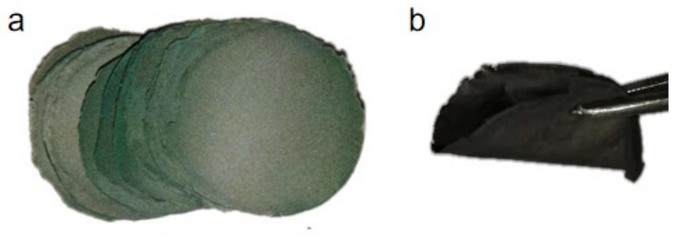

Fig. S6 Physical display of (a) GCu films and (b) GCu600 films.

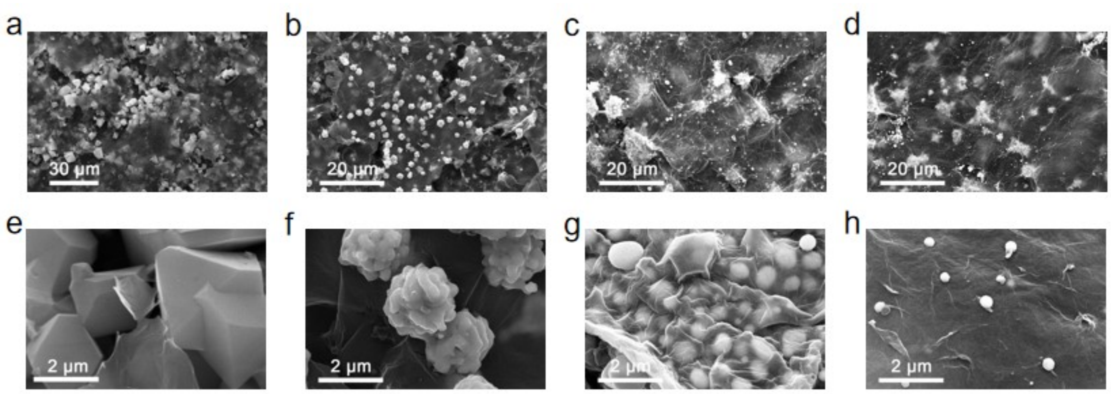

Fig. S7 SEM images of the (a and e) GO/HKUST-1, (b and f) GCu400, (c and g) GCu600 and (d and h) GCu800 samples.

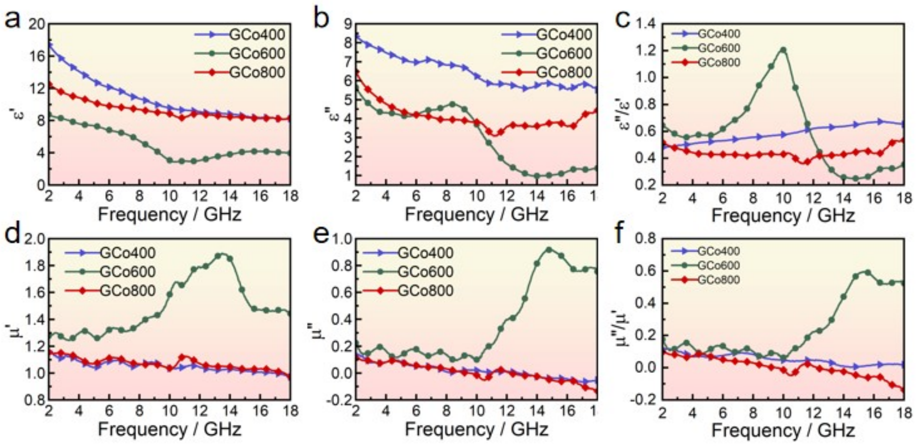

Fig. S8 (a, b, d and e) EM parameter and (c and f) loss tangent of GCo samples.

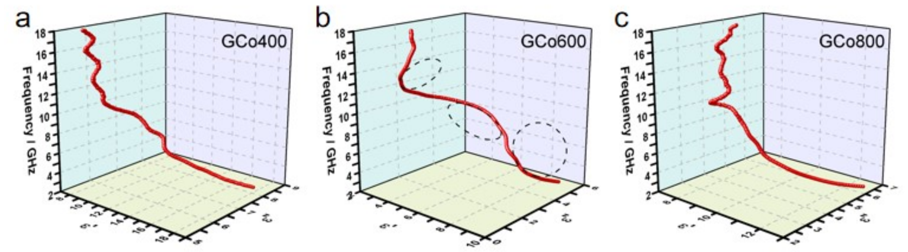

Fig. S9 Cole-Cole curves of GCo samples.

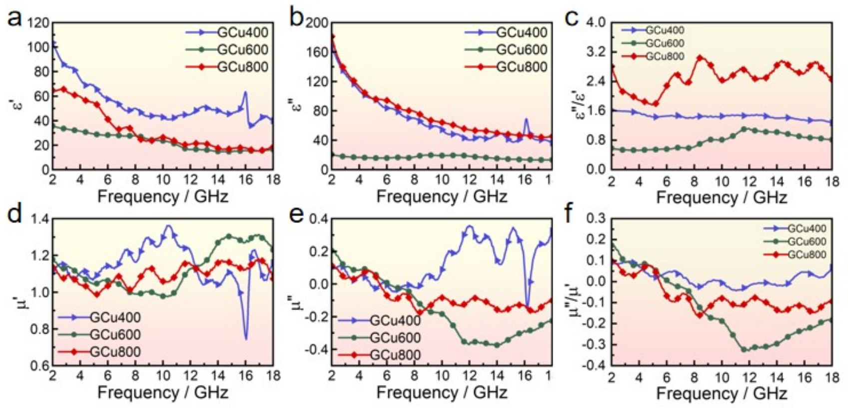

Fig. S10 (a, b, d and e) EM parameter and (c and f) loss tangent of GCo samples.

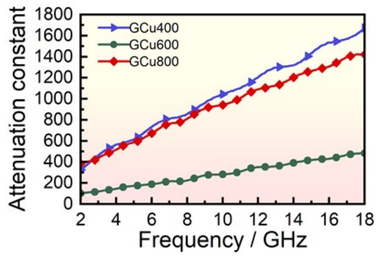

Fig. S11 Attenuation constant of GCu samples.

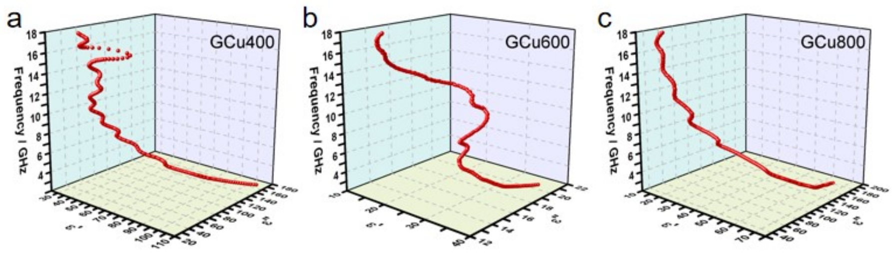

Fig. S12 Cole-Cole curves of GCu samples.

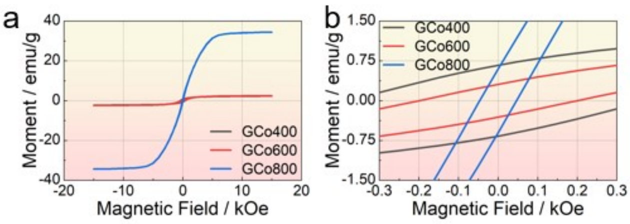

Fig. S13 Magnetic hysteresis loops of GCo600.

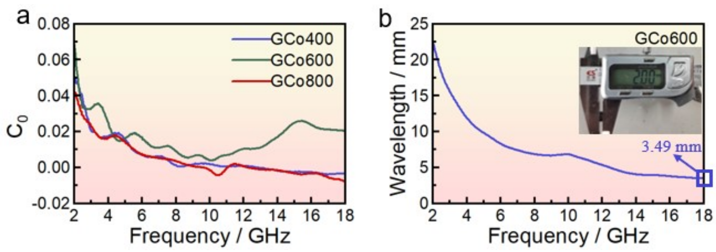

Fig. S14 C_0 curves and frequency dependence of EM wave transmitting in testing sample.

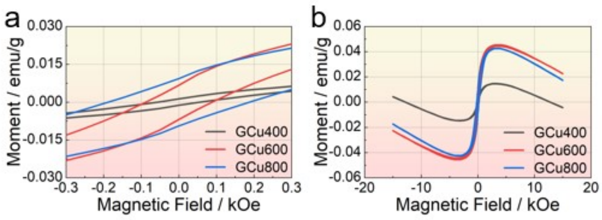

Fig. S15 Magnetic hysteresis loops of GCu600 samples.

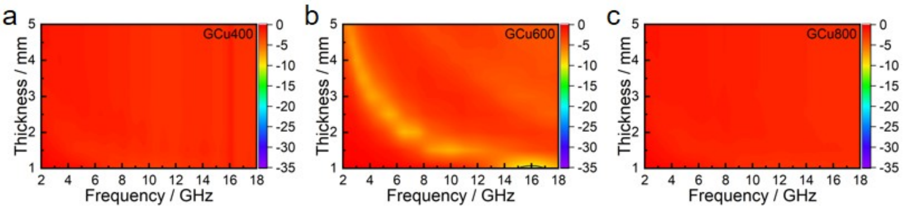

Fig. S16 Frequency dependence of contour maps of (a) GCu400, (b) GCu600 and (c) GCu800 samples at different thicknesses.

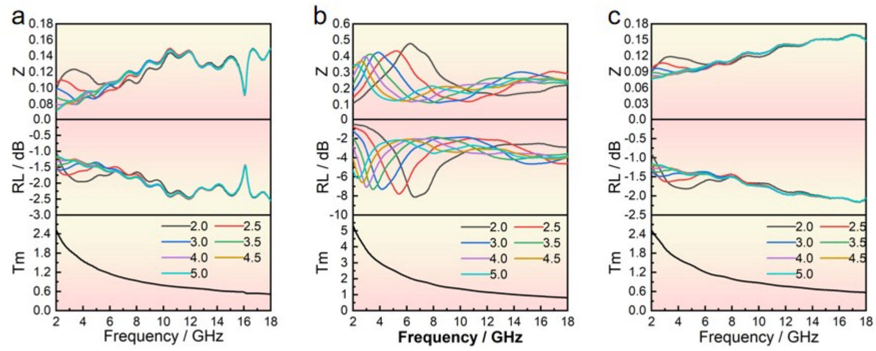

Fig. S17 Comparison between *RL*, *Z* and T_m of (a) GCu400, (b) GCu600 and (c) GCu800 samples.

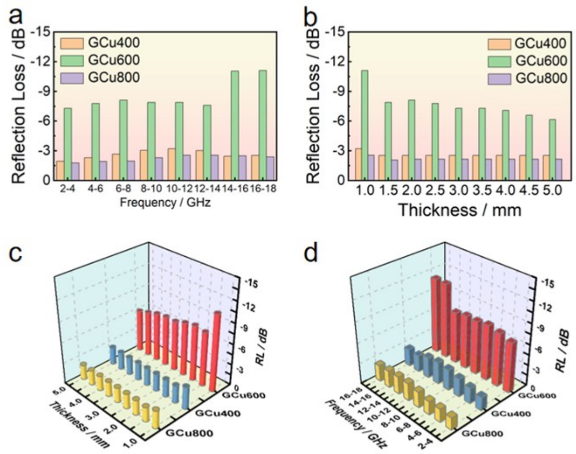

Fig. S18 Comparison of *RL* values at different (a and c) frequency and (b and d) thickness of GCu samples.