Supporting Information

Hong Lei,^{a,b} Zhuo Liu,^a Chong He,^{a,b} Shou-Chun Zhang,^a Ye-Qun Liu,^a Cheng-Jie Hua,^{a,c} Xiao-Ming Li^a, Feng-Li,^{a,b} Cheng-Meng Chen,^{*a} and Rong Cai^{a,d}

a Key Laboratory of Carbon Materials, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan 030001, P. R. China

b University of Chinese Academy of sciences, Beijing 100049, P. R. China

c School of chemical engineering and technology, China University of Mining & Technology, Xuzhou 221116

d Academy of Opto-Electronics, Chinese Academy of Sciences, Beijing 100094, P. R. China

E-mail: ccm@sxicc.ac.cn



Fig.S1 The C1s XPS spectrum of graphene.

Our previous reference report that after thermal reduction, part of the oxygencontaining functional groups were removed. The summary of XPS fitting results, which quantitate the relative abundance of each species, were listed in Table S1. C1 (284.5eV), C2(285.6eV), C3(287.9eV), C4(289.3eV), C5(291.4 eV) components were assigned to C-C, C-O, C=O, C(O)O, related groups and graphitic shake-up satellites, respectively.

B.E(eV)	C1(284.5)	C2(285.6)	C3(287.9)	C4(289.0)	C5(291.4)
					Graphtic shake-
Assignment	C-C	C-0	C=0	C(O)O	up

Table S1 fitted results (%) of C1s XPS spectra of graphene



Fig.S2 showed TGA curves of neat LDPE and LPGNs with different graphene content.

TGA plots can obtain temperature of the extrapolated onset (T_e) with air, they were 357.4 °C, 407.9 °C, 408.5 °C and 415.4 °C with neat LDPE and 0.2wt%, 0.5wt%, 0.8wt% of graphene, respectively.



Fig.S3 Tensile curves of LDPE and LDPE-0.8wt%GN with different processing.

LDPE(nopre)-0.8wt%GN represented LDPE-0.8wt%GN didn't take measure preprocess procedure which just simple mixed before melt compounding. Fig.S3 showed tensile curves of neat LDPE and LDPE-0.8wt%GN with different processing method. The mechanical properties of LDPE-0.8wt%GN without premixing was very poor, because inhomogeneous distribution of graphene caused a large number of defects in the LEPE matrix.



Fig.S4 (a) XRD patterns of neat LDPE and LDPE-0.8wt%GN with different processing. (b) TGA curves of neat LDPE and LDPE-0.8wt%GN with different processing.

Reaggregation of graphene is a question that must be considered in nanocomposites. In order to prevent reaggregation of graphene, our experimental procedure was continuous without any delay. We took a more extreme case to illustrate the effect of graphene aggregation on the performance of LDPE. Crystallinity and thermal properties of LDPE(nopre)-0.8wt%GN researched by XRD and TGA. Specific results can be seen Fig.S4a and Fig.S4b. Graphene agglomeration as impurities cannot provide heterogeneous nucleation sites for polymer crystallization. From extreme case we can know that graphene suspension reaggregation will reduce the effect of increasing the crystallinity. Thermal stability of LDPE (nopre) -0.8wt%GN was worse than LDPE-0.8wt%GN, but it better than neat LDPE. The presence of graphene can reduce oxygen diffusion and capture some free radicals. Dispersed graphene as small rooms separated the alkyl radicals, and many radicals became relatively isolated in LPGNs. Graphene dispersed inhomogeneous in matrix which will significantly reduce the thermal stability of the LPGN-0.8wt%GN.

Strain-aging phenomenon has been researched in the composites. It was intriguing that the composites did have a strain-aging phenomenon. LDPE-0.2wt%GN was used to do tensile test until the stress reached the yield point, and then stopping the experiment. The sample after the test was placed in an oven for 10 hours at 100 °C. When the sample cooled, second tensile test was done. The second yield strength was

higher than the first one which illustrate that the composites had strain-aging phenomenon. Fig.S5 showed the difference between the specific results of the two experiments. The composites occurred strain-aging phenomenon because the first stretch made the crystal orientation of LDPE-0.2wt%GN more obvious and the change of interactions between graphene and molecular chain.



Fig.S5 Strain-aging phenomenon of LDPE-0.2wt%GN.

Fatigue data was very important for evaluation of materials. Cycle times (N) were the average of the three tests for every sample. And then take logarithm of N that was lgN. N and lgN were presented in more detail in table S2 and table S3 as follows.

Table S2 N of samples						
Samples	σ_{max} /MPa					
Samples	9	6	4			
LDPE-0wt%GN	120	2347	3266			
LDPE-0.2wt%GN	223	2832	3532			
LDPE-0.5wt%GN	538	2533	3774			
LDPE-0.8wt%GN	706	3401	5608			
Table S3lgN of samples						
Complex	σ _{max} /MPa					
Samples	9	6	4			
LDPE-0wt%GN	2.08	3.37	3.51			
LDPE-0.2wt%GN	2.35	3.45	3.55			
LDPE-0.5wt%GN	2.73	3.40	3.58			
LDPE-0.8wt%GN	2.85	3.53	3.75			