Supplementary Information

Sandwich-like Graphene Nanocomposites Armed with Nanoneedles

Bora Nam, Ha-Jin Lee, Hyeah Goh, Young Boo Lee, and Won San Choi
Fig. S1 a) XRD and b) TEM/EDX analyses of GNS/Au-4/IO. The XRD and EDX analyses confirmed synthesis of gold and hematite, indicating formation of the GNS/Au/IO. The H is denoted as hematite. The Cu peaks came from the Cu grid.
**Fig. S2** TEM/EDX analysis of GNS/Au-4/IO/SiO$_2$. The EDX analysis confirmed formation of the SiO$_2$ layer on the GNS/Au/IO. The Cu peaks came from the Cu grid.
Fig. S3  a) SEM and b) STEM images of GNS/Au/IO-4/SiO$_2$. 
**Fig. S4** SEM images of a) GNS/IO-1 and b) GNS/IO-5. The GNS/IO-5 is clearly distinguishable from the GNS/IO-1 due to the long needles of iron oxide. The specific surface areas of the GNS/IO-1 and the GNS/IO-5 were found to be 655 and 1270 m$^2$/g, respectively.
Fig. S5 Time-dependent UV-vis absorption spectral changes of 4-NP reduction by the GNS.

The GNS showed extremely lower catalytic properties, and the reaction did not finish. It did not show comparable performance for nanocatalysts (GNS/Au, GNS/Au/IO, and GNS/Au/IO/SiO₂).
**Fig. S6** Time-dependent UV-vis absorption spectral changes of 4-NP reduction by NaBH$_4$ in the absence of nanocatalysts. The reaction did not proceed even in the presence of a large excess of NaBH$_4$. 
Fig. S7 STEM images of the a) GNS and b) GNS/IOs. c) Time-dependent UV-vis absorption spectral changes of 4-NP reduction in the presence of the GNS/IOs without AuNPs. The GNS/IOs showed extremely lower catalytic properties, and the reaction did not finish.
Fig. S8 UV-vis absorption spectral changes showing adsorption of a) 4-NP or b) 4-AP on the GNS. Carboxylic acid or PSS groups on GNSs are able to react with ionic species (amine groups of the 4-APs) and form precipitates. Apparently the reactive surface of the GNSs was irreparably damaged by repeated adsorption and desorption of ionic species.
Fig. S9 Catalytic recycling ability of GNS/Au-4/IO for a fifth reaction. It showed relatively good catalytic performance until the fourth reaction. However, thereafter, its performance rapidly decreased until the reaction failed to finish owing to decreasing stability of GNC via either an ion exchange reaction or the formation of strong complexes. After 1350s, the reaction did not proceed anymore (until 20 h).
Fig. S10 Time-dependent UV-vis absorption spectral changes of 4-NP reduction in the presence of the GNS/Au-4/IO/SiO$_2$ (without needle-like coating). It showed lower catalytic properties.