



High Efficiency Graphene Oxide Catalysts for Oxidative Reactions

Market Need

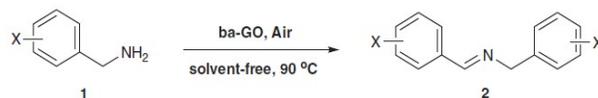
In the drive towards green and sustainable chemistry, a constant refrain is the use of non-toxic, sustainable catalysts with minimal environmental footprint. The use of carbon catalysts is attractive in this regard because of their low cost and natural abundance.

Graphene oxide sheets, inexpensively prepared from graphite, have been used in various chemical reactions as catalysts in place of expensive metal catalysts. However, they typically have very low catalytic reactivity and require a catalyst loading of 200-400 wt% to substantially convert a reactant to a desired product. This low reactivity limits widespread use of graphene oxide catalysts. Hence, there is an urgent need to increase the catalytic efficiency of graphene oxide catalyst in order to make it a viable option for industrial use.

Solution

Prof. Loh Kian Ping's group from the Department of Chemistry has developed a highly efficient graphene oxide catalyst (abbreviated as ba-GO) by the chemical treatment of conventionally synthesized graphene oxide. Due to the enhanced catalytic efficiency, only a very small amount of catalyst loading (less than 5 weight %) is needed during catalytic reactions. This represents a breakthrough with important applications in chemical industry requiring the use of metal catalysts, as our invention provides a new type of low cost, environmentally friendly carbon catalyst that can substitute the metal catalysts used in the industry.

As an example, oxidative coupling of amines to imines under solvent free, open air conditions have been achieved with yields up to 98% and low catalyst loading (5 wt%). Table 1 below illustrates the oxidation of benzylamines using ba-GO as catalyst. The reusability of ba-GO as a catalyst in benzylamine oxidation reaction is illustrated in Fig. 1, demonstrating a high yield of 93% at the sixth cycle. Our studies have been published in Nature Communications. Our office has also filed for patent application for this invention.

Table 1: Oxidation of benzylamines using ba-GO₃ catalyst

Entry	Substrate (Compound No)	Product (Compound No)	Reaction Time (hours)	Yield (%)
1	X = H (1a)	X = H (2a)	12	98 ^a
2	X = H (1a)	X = H (2a)	12 (6 th cycle)	93 ^a
3	X = <i>p</i> -Cl (1b)	X = <i>p</i> -Cl (2b)	12	96 ^b
4	X = 1,2-diCl (1c)	X = 1,2-diCl (2c)	12	94 ^b
5	X = <i>p</i> -Me (1d)	X = <i>p</i> -Me (2d)	11	94 ^b
6	X = <i>m</i> -Me (1e)	X = <i>m</i> -Me (2e)	12	92 ^b
7	X = <i>o</i> -Me (1f)	X = <i>o</i> -Me (2f)	13	95 ^b

^a Yield calculated using GC with anisole as the internal standard.
^b Isolation yield.

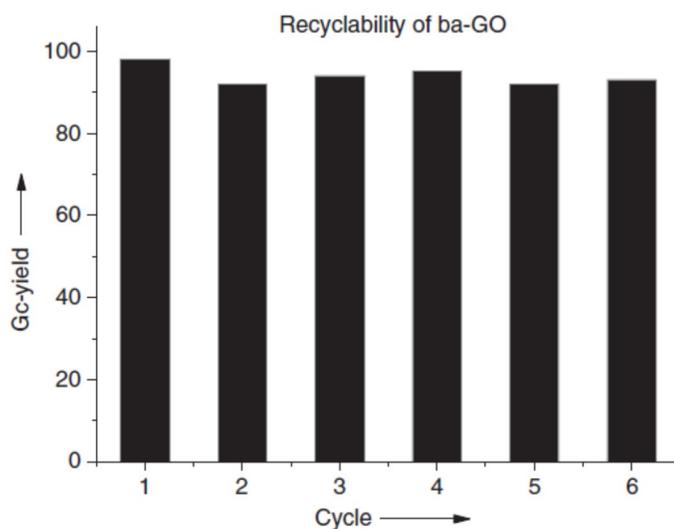


Fig. 1: Reusability of ba-GO as a catalyst in benzylamine oxidation reaction. Conditions: 90 C, open air, 1.0 g PhCH₂NH₂, 50 mg catalyst (5 wt%), 12h. It was reused as a catalyst for 6 cycles. Unexpectedly, at the sixth cycle, it was still capable of converting benzylamine to N-benzylidene benzylamine in a yield as high as 93%.

Application and advantages

Potential application is as a catalyst in chemical synthesis or transformation, especially for oxidative reactions. For example, the catalyst may be used in pharmaceutical applications eg. anti-inflammatory, anti-cancer, anti-bacterial and anti-fungal.

The advantages are:

- Ba-GO is a low-cost and abundant alternative to expensive metal catalyst
- More than 40X improvement in catalyst loading as compared to untreated graphene oxide catalyst
- High catalytic efficiency (demonstrated up to 98% yield)
- Reusable (demonstrated reusability with high yield (93%) after repeated use (6th use))

Keywords

Graphene oxide, catalyst, oxidation

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