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Graphene opens pathways to a carbon-neutral cement industry

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Global cement production has increased 30 times since 1950 and nearly 4 times since 1990, becoming the third-largest source of anthropogenic carbon dioxide (CO_2) emissions after fossil fuels and land-use changes [1]. But the cement industry is under scrutiny. In order to achieve the Paris Agreement's targets and reach net-zero CO_2 emissions by 2050, it needs to cut its emissions from the current 2.2 to ~1.5 gigatonnes of CO_2 per year within the next decades (Fig. 1a) [2].

However, achieving carbon neutrality in the cement industry is challenging due to its intrinsic combustion or process emissions. Producing 1 tonne of clinker, Portland cement's main component, directly releases about ~0.86 tonnes of CO_2 into the atmosphere (i.e., carbon emission coefficient ~0.86). About 60% of these emissions are generated from the conversion of limestone ($CaCO_3$) into lime (CaO) during the cement burning or clinkering stage [3]. The other 40% is mostly associated to the burning of fossil fuels required to reach the clinkerization temperature (~1450 °C). Emissions from the heat treatment of raw materials may also increase the carbon footprint. The carbon emission coefficient of the cement industry is not high compared to that of other structural materials (e.g., carbon emission coefficient of steel production ~1.9). But given that Portland cement is the most consumed manufactured material in the world, and that the cement industry alone is responsible for 5% of global CO_2 emissions [2], further reduction in carbon emission coefficient is expected by the Paris Agreement.

Several strategies have been proposed to achieve carbon neutrality in the cement industry [2,4,5], including: (1) improving the energy of the cement production process (e.g., reducing kiln heat losses); (2) increasing the use of alternative fuels; (3) promotion of novel manufacturing technologies for cement and concrete production (e.g., carbon capture and storage); (4) blending cement with industrial by-products; and (5) development of clinker-free cements (e.g., geopolymer) and cements with less clinker (e.g., limestone calcined clay cement). Most of these have been slowly driven forward because of technological difficulties, material shortages, or high costs. Besides, it is currently impractical to reduce the demand for cement because of continued global urbanization. In fact, the global annual concrete consumption of 25 billion/year is expected to increase by 12%–23% by 2050 [6]. Therefore, it is imperative to develop immediate solutions to reduce CO_2 emissions from both the cement industry and construction sectors.

Recently, graphene-based nanosheets (GNS), including graphene, graphene oxide (GO), reduced GO (rGO), and graphene nanoplatelets (GNP), have attracted attention as a promising candidate to enhance the physicochemical properties of cementitious materials. Due to their superior mechanical and impermeability properties, combined with a densification effect on the cement microstructure, very small addition of GNS (as small as 0.01%–0.05% of the weight of the Portland cement) to cementitious materials can render composites with significant enhancements in compressive/tensile strength (>80% increase) and durability (e.g., >500% improvement in water penetration resistance) [7,8]. Furthermore, the excellent electrical and thermal conductivity of GNS (particularly of graphene and GNP) can endow cementitious materials with smart properties such as self-sensing and thermal efficiency [7]. Now, state-of-art research also shows that GNS-reinforcement can be a cost-effective strategy to reduce the environmental impact of concrete production and maintenance, in addition to improving the efficiency of building construction and operations—to provide an effective approach to reducing CO_2 emissions and realizing carbon neutrality in the cement and construction sectors.

Accordingly, we identified six potential pathways by which the application of GNS can efficiently bolster the development of sustainable construction materials from the carbon-neutral perspective (Fig. 1b). These strategies can be developed and implemented concurrently to fully deliver target environmental benefits of GNS-reinforced concrete technology in the construction field.

(i) *Reducing the consumption of Portland cement/concrete.* GNS can deliver improvements to the compressive strength of cementitious materials by up to 150%, in addition to increasing the Young's modulus by a figure of 80% [7]. Such significant mechanical enhancement can be exploited to design structures with smaller cross-sections or reduced Portland cement content in the concrete mix, which would be directly converted into CO_2 savings. For example, Dimov et al. [7] pointed out that if we could achieve a 50% reduction in the required Portland cement by using GNS while still fulfilling the specification for the loading of buildings, 0.45 tonnes of CO_2 eq. per tonne of manufactured cement would be saved. In a more complete lifecycle assessment, Papaniko-

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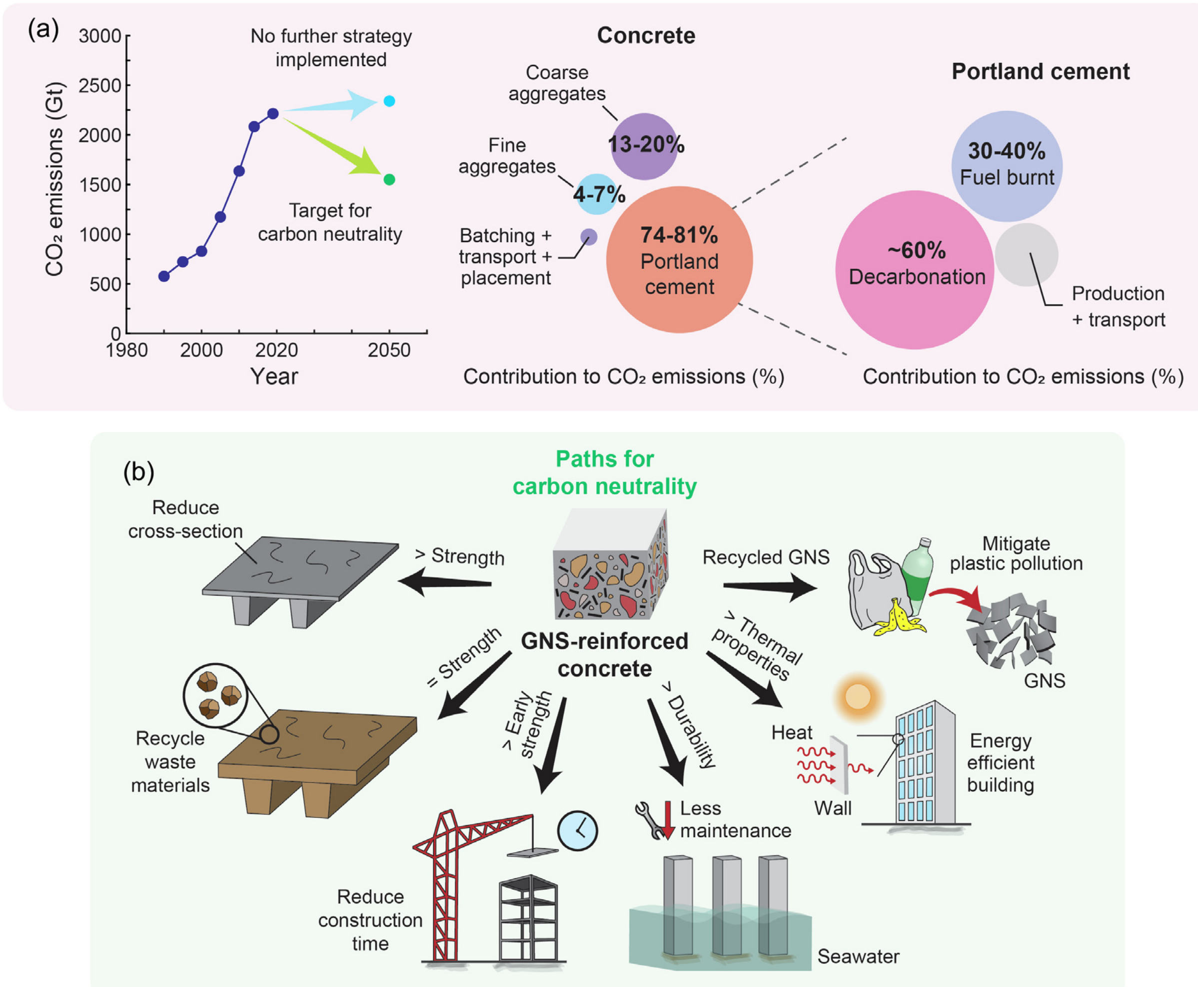

Fig. 1. Potential pathways by which the application of graphene-based nanosheets (GNS)-concrete materials can lead the cement industry to net zero emissions by 2050. (a) Absolute CO_2 emissions by the cement industry per year from 1990 to 2014 [1]. The figure also shows the predicted CO_2 emissions in 2050 in case effective mitigation strategies are implemented or not [2,6]. Portland cement manufacturing contributes to most of the CO_2 emissions associated with concrete materials [4]. Within Portland cement manufacturing, the decarbonation process is responsible for more than half of the CO_2 emissions [3]. (b) Schematic of the six practical strategies to achieve carbon-neutral construction materials with GNS.(For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

laou et al. [9] recently analysed the environmental footprint of GNP-reinforced concrete, including the impacts from resource extraction to the factory gate during GNP production. They demonstrated that the production of 1 kg of GNP results in 0.17 kg of CO_2, compared with 0.86 kg CO_2 for Portland cement, validating the environmental benefits of the nanomaterial. Their sensitivity analysis also disclosed that if the addition of GNP results in a 5% reduction in the amount of Portland cement in a concrete mix, the impact of manufactured concrete materials on global warming would be reduced by ~20%. Further research is needed, however, to more fully evaluate the environmental impact of concrete reinforced by GO. On one hand, the fabrication of monolayer GO requires more chemical processing steps compared with that of multi-layer GNP. On the other hand, less GO is needed to achieve similar improvements in strength and durability.

(ii) *Increasing the utilization of by-products/waste materials in concrete.* The increase in strength provided by GNS can also enable the use of by-products or marginal recycled materials that are often detrimental to the mechanical properties of concrete. This approach will not only reduce the CO_2 emissions from the cement industry but also assist in mitigating the environmental impact of other industrial sectors. For example, Long et al. [10] used GO to improve the mechanical properties of mortars using construction and demolition waste, such as recycled fine aggregates, which can alleviate the environmental burden of both natural aggregate extraction and landfill utilization. Their lifecycle assessment indicated that, for an equivalent mechanical strength, the GO-reinforced mortar with recycled fine aggregate can potentially reduce the associated greenhouse gas emissions by ~7% compared with a reference mortar with natural aggregate. In addition, the process of fabricating the GO-reinforced mortars would consume less energy (a reduction of ~2.2%). On the other hand, we note the importance of considering in future research the long-term durability in the lifecycle assessment of waste-incorporated concrete materials.

(iii) *Reducing construction costs.* Interestingly, it has been also demonstrated that GNS can reduce the cost of construction materials either due to the improvements in strength or the increased incorporation of by-products/waste materials. For instance, Devi and Khan [11] demonstrated that concrete materials with the addition of 0.02 wt%–0.08 wt% of GO can be cost-effective. Their cost analysis indicated that, although the total cost of the GO-reinforced concrete was ~2%–7% higher than for the reference mix, the economy index (compressive strength/cost per m^3) of the GO-reinforced mixes was increased by 25%–40%. Economical GNS-reinforced fly ash cement composite made with fine recycled aggregate has been also demonstrated by Sharma and Arora [12]. As the GNS addition outweighed the mechanical property loss from the incorporation of recycled aggregate, the cost of the GNS-reinforced mixtures was ~35% lower compared with the reference mortar with natural aggregate and the same compressive strength. Other studies also found that GNS can significantly accelerate the strength development of concrete [13], and thereby potentially reduce construction times/labour costs.

(iv) *Reducing maintenance/repair/renovation costs.* The effect of GNS on the durability of cementitious materials is even more pronounced than on the strength, with up to 500% improvement in certain durability properties [7]. In addition, GNS can impart self-sensing properties to cement composites, which can assist in structural health-monitoring of infrastructure. These outcomes indicate that GNS-reinforcement has great potential to prolong the service life of concrete and contribute to more durable infrastructure. This, in turn, can lower CO_2 emissions via a reduction in the amount of construction materials and energy consumed that are associated with maintenance, repair, and renovation of infrastructure. Previous lifecycle assessments of GNS-reinforced cementitious materials have not considered their service-life durability, and further research is needed in this area.

(v) *Creating functional, energy-efficient materials.* Building operations, including heating and/or cooling, account for a significant share of energy-related CO_2 emissions worldwide, creating a need for the development of energy-efficient buildings. Novel cementitious composites with enhanced thermal properties via GNS reinforcement remain prospective, opening the door to sustainable energy-efficient concrete structures. For instance, GNS delivers improved effectiveness of thermal energy storage construction materials [14], a promising class of materials with application in the thermal regulation of buildings, which can be directly translated into savings in the energy consumed in heating, ventilation and air conditioning.

(vi) *Using GNS from plastic waste.* Given the large annual concrete production (>20 billion tonnes), incorporating waste materials into concrete is one of the most effective methods to reduce the impact of both types of material on the environment. Nevertheless, mixing waste plastics into concrete is still impractical due to the degradation in mechanical and durability properties. Fortunately, the recent discovery of a flash Joule heating method to synthesize graphene from waste carbon sources, such as discarded food, rubber tyres, and mixed plastic waste [15] opened a potential route to mitigate the immediate crisis of waste plastics, of which more than 200 million tonnes are generated every year. Luong et al. [15] already demonstrated that the addition of flash graphene into Portland cement paste can improve its compressive strength, and further research in this area poses a great opportunity for the recycling sector to mitigate the global impact of both plastic and cement industries.

In summary, the improved performance of concrete by GNS reinforcement not only enables the fabrication of cost-effective, high-performance concrete for skyscrapers or cross-sea bridges, reducing their maintenance and renovation, but also can contribute to the development of modern construction with energy efficiency and intelligence, all of which play a crucial role in attaining carbon neutrality in the cement industry and construction sectors. Moreover, by enabling the re-utilization of by-products/waste materials such as recycled aggregates and plastics, further development of GNS-reinforced concrete materials can potentially augment carbon neutrality targets of other industry sectors such as mining, oil, and gas.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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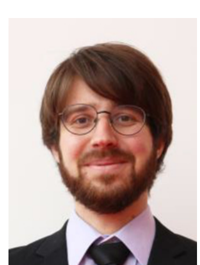

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