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Leveraging engineered nanomaterials to support material circularity†

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Materials are central to a circular economy. They enter one of two cycles, the biological (where materials serve a purpose and then degrade as nutrients for renewing natural resources) or the technical (where materials perpetuate in products, serving a single or multiple functions with each subsequent use). Given the ubiquity of engineered nanomaterials in products supporting nearly every facet of society, it is critical to consider how they integrate in these circular resource flows. In this perspective, we present an historical overview of the emergence and advancements of engineered nanomaterials as well as provide context for identifying tractable avenues for them to advance the circular materials economy. We highlight the opportunity for engineered nanomaterials to improve the performance and extend the life of bulk composite materials. We outline a specific example of nano-enabled concrete to demonstrate the opportunity and elucidate the importance of low additive amounts, high functional gain, and low additional embodied energy of the nanomaterial used. Finally, we offer perspective on future opportunities for ongoing research in our field to support successful realization of a global circular materials economy.

Environmental significance

Engineered nanomaterials are now incorporated in wide ranging resource flows and into products across global markets. Thus, as society transitions from linear to circular material systems, it is not a question of whether engineered nanomaterials will be a part of that transition, rather it is determining what role they can play to facilitate and enhance the circular materials economy.

A brief orientation to circular resource flows

Over the past half-century, engineered nanomaterials have evolved technologically from concept to reality through research and development in academic and industrial labs. Engineered nanomaterials fuel an impressive $68 billion global nanotechnology market (2023 data), which is expected to reach $184 billion by 2028.[1] Numerous engineered nanomaterials are now produced commercially and incorporated into a wide range of products, giving rise to a vast array of nano-enabled products distributed across the global material ecosystem. Healthcare, pharmaceuticals, electronics, and semiconductor applications make up over half the global market share, while the increased adoption of nanotechnology in the energy and agriculture sectors are identified as key market drivers.[1] With increased market penetration and diversity of end uses, it is important to consider what the incorporation of engineered nanomaterials means for the end-of-life management of these products, as well as to identify opportunities for their incorporation to enhance the transition away from the current linear to circular resource systems. In this perspective, we explore the direct and (perhaps more important) indirect mechanisms by which engineered nanomaterials can support a circular materials economy.

In an ideal world, products would be long-lasting, repairable, and designed to readily reuse or disassemble for recycling, with associated material recovery. However, this is far from what happens for most products in practice. End-of-life recycling rates for most elements and materials remain abysmally low. For example, on a mass basis only 35% of municipal solid wastes are recycled,[2] and more than half of metals assessed have recovery rates of <1%.[3] There is

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widespread acknowledgement that the largely linear system of production has contributed to large human inequities and environmental damage to the planet and must evolve into one that can be sustained in the long term, where an equal amount of the ingenuity and engineering that goes into creating materials and products also goes into recovering them to use again.

The circular economy has emerged as a popular framework for envisioning how a transition to a more sustainable materials management system might be achieved. In a circular economy, bio-based materials are created from renewable resources and designed to degrade safely in a biological cycle, while finite, abiotic resources are used efficiently and cycled repeatedly in a technical cycle.[4] Further, a circular business model can be defined as a business model for value creation based on economic value retained in products after use.[5] Historically, many business models have not formally considered the circular economy, although circular resource loops including recycling, downcycling, reuse, repair, and remanufacturing have existed for considerable periods.[6] Newer models increasingly make use of diverse strategies for preserving valuable resources and exhibit greater sophistication. The Organization for Economic Co-operation and Development (OECD) outlines five genres of circular business models : (1) circular supply, (2) resource recovery, (3) product life extension, (4) sharing, and (5) product service system.[7] This elegant vision of closed cycles and zero waste has inspired many governments and companies to develop circular economy metrics, goals, and economic incentives, but there remain practical, technical, and economic challenges to fully realize these for harmonized global implementation.

Opportunities for engineered nanomaterials in the transition to circular resource systems

In a recent perspective, Gilbertson and Vikesland[8] outline various opportunities for engineered nanomaterials in a circular economy. These strategies include (1) designing out inherent hazards based on advances in structure–property-hazard guidelines (2) generating engineered nanomaterials from bio-based and renewable feedstocks, (3) programing into their initial design the ability to adapt to multiple uses, and (4) enhanced separation capabilities from complex matrices based on inherent properties. Such opportunities focus on engineered nanomaterials themselves. We envision many additional opportunities (and trade-offs) when circular economy concepts are applied across multiple materials,[9] as is the case for most products. In this perspective we ask: how might engineered nanomaterials help or hinder a circular economy?

One may first address this question by considering the direct recycling and recovery of engineered nanomaterials themselves, but in many ways, this is the least developed and most technologically challenging loop to consider. Most often, engineered nanomaterials are one component of a material or product, and recycling addresses a product at the end of life when the functionality has diminished. Engineered nanomaterials are much more energy-intensive to produce than bulk materials and the ability to reuse expensive or critical materials[10,11] such as indium or gallium, suggests that their recovery would have environmental benefits if substituted for newly synthesized nanomaterials. However, actually recovering these nanomaterials from end-of-life products is difficult for a number of reasons. First, engineered nanomaterials are typically present in products in very low concentrations, while recycling processes are likely to be optimized around the bulk materials. There is also a thermodynamic argument that the energy required to separate and purify engineered nanomaterials present in low concentrations may negate the environmental benefits of their recycling.[12] But for nano-waste streams that are relatively uniform with high concentrations of valuable materials (*e.g.*, gold, platinum group metals) or critical elements (*e.g.*, nickel, cobalt) in applications such as spent industrial process or automotive catalysts or used electric vehicle batteries, recovery may be economically and environmentally favorable.[13] Pyro- or hydrometallurgical recovery and processing of nano-waste streams may destroy the nanostructures requiring reformation (potentially *in situ*).[14]

Second, engineered nanomaterials make up a very small portion of the materials market by mass, comprising 2% of global cerium and 1% silver, but $\ll$1% for most other engineered nanomaterials types.[15] Recycling technologies often employ techniques that are element- or material-specific, such as magnetic separation for ferrous materials, and in theory the same techniques used for bulk materials could be used to recover engineered nanomaterials, as has been demonstrated for magnetic nanomaterials.[16] Direct recycling could make economic sense for applications where waste nanomaterials exist in high concentrations, such as recycling carbon black from tires. But for mixed material flows with low-value nanomaterials, it makes more economic sense for recyclers to prioritize the larger flows of used products and scrap that make up most of the market in mass terms.

Third, several engineered nanomaterial applications are designed to be inherently dissipative in the environment, such as engineered nanomaterials used in agriculture, making collection and recovery a practical impossibility. In the cases where the engineered nanomaterial is not released, it may transform in the product during use. In such cases where this transformation results in elimination of the desirable property(ies), the nanomaterial becomes less desirable to recover. Finally, engineered nanomaterials are often tightly bound in composite material matrices such that the processes required for separation such as thermal decomposition or acid digestion can change the morphology or surface chemistry of the engineered nanomaterials, rendering them useless if those properties underlie their intended function.

Even in cases where improving the circularity of engineered nanomaterials themselves is technologically challenging and/or environmentally questionable, there are other major leverage points to enable the circular economy. One approach that is gaining traction and demonstrated success is the use of engineered nanomaterials to enable or improve element recovery in separation and purification processes, for example for recovery of rare earth elements from dilute waste streams.[17,18] In absolute mass terms, however, the opportunity that we believe is most important is harnessing engineered nanomaterials to improve the performance of bulk materials, leading to more material-efficient designs and/or longer-lived products, with overall reductions in material demand and associated embodied energy and emissions.

Integrating nanomaterials to enable circular bulk materials: greater functional gain with less material

To date, engineered nanomaterials have been incorporated into many bulk materials, introducing advantageous cascading benefits, such as in (1) lightweight, high strength composites in transportation (benefits: greater fuel efficiency, reduced repair/maintenance to vehicle body), (2) food packaging (benefits: food preservation and protection that reduces food waste), and (3) antimicrobial textiles and polymer composites (benefits: infection prevention, reduce/eliminate microbial transmittance). Integrating existing sustainable design and circular economy frameworks will ensure growth in nanocomposites that benefits rather than diminishes natural resources.

Nanoscience and engineering has advanced the ability to control engineered nanomaterial characteristics through manipulating parameters during and immediately following synthesis (*e.g.*, varying the reagents, solvent, temperature, capping ligands).[19–21] As a result, engineered nanomaterials can be designed with specific size, surface chemistry, and shape, which in turn controls functional properties such as surface charge, surface reactivity, optical absorbance and emissions, electrical conductivity, and semi-conductivity. Example applications include manipulating quantum dot size to induce discrete transitions in the optical emission, ranging from blue (smaller particle size) to red (larger particle size), to enable unprecedented resolution in applications from display screens to medical imaging.[22,23] Similarly, the manipulation of nano-silver particle size and shape controls the surface oxidation potential and thus, the production of silver ions,[24] which are used in wide ranging antimicrobial applications. This tunable technology can serve as an alternative to troublesome chemical alternatives (*e.g.*, triclosan) and conventional antibiotics, while having the potential to elude or delay antimicrobial resistance without compromising efficacy.[25] The ability to manipulate functional properties of engineered nanomaterials, in conjunction with reducing adverse impacts that emerge from synthesis and exposure, has been enabled through the integration of the 12 Principles of Green Chemistry in synthesis design.[26–29] Achieving these dual objectives – enhanced functional performance and reduced adverse impacts – is a key way in which engineered nanomaterials are enabling a circular resource economy.

Material efficiency[9] encompasses gaining function in a material in an additive manner (Fig. 1). Function can be gained by adding small weight percentages of engineered nanomaterials to a bulk material system. This has the potential to improve the functional performance of the composite material with little added mass in comparison to achieving the same improvements through substantial addition of non-nano or bulk materials. Since the mass of any material added is directly proportional to the magnitude of its environmental impact from manufacturing, achieving greater functional performance with lower mass quantities can reduce environmental impacts, as long as the manufacturing is not overly resource- or emissions-intensive. Examples where small amounts of engineered nanomaterials (*e.g.*, 0.01–5% wt) introduce large gains in performance include (1) the use nano-clay additives (*e.g.*, montmorillonite, bentonite) in composites for uses ranging from packaging to automotive parts (note: clay nanocomposites consistently account for over 50% of total global nanocomposite market share[30]). A range of engineered nanomaterial compositions are used to add conductivity to polymer composites for applications ranging from wearable electronics to

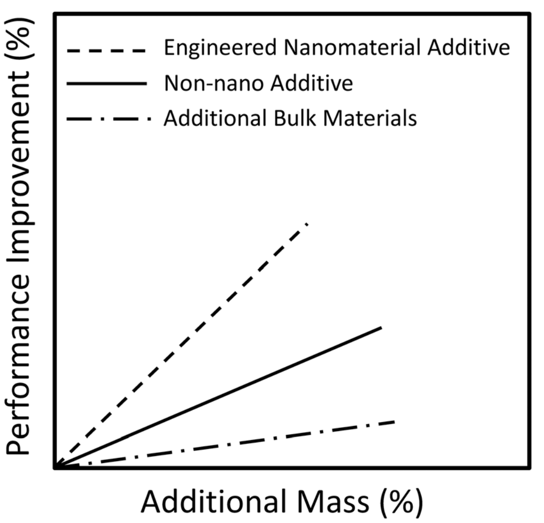

Fig. 1 Material efficiency is the concept in which the ratio of functional gain to material mass increases compared with a baseline, alternative, or *status quo*. This figure illustrates the concept for engineered nanomaterials wherein the gain in function of the bulk material per unit nanomaterial mass added is much larger than for non-nanomaterial approaches. The slope indicates the material efficiency and the linear trends are demonstrative.

photovoltaics.[31] Engineered nanomaterials are also enabling structural materials, enhancing durability and strength properties advantageous for reducing deterioration and improving performance in roads, bridges, and buildings. One example of this is nano-enabled concrete, discussed in the following section.

To realize the opportunity for high performing, material efficient, nano-enabled bulk composite materials, it is essential that the impact associated with the added nanomaterial is not greater than the gain in functional performance or extended lifetime of a bulk or composite material.[32–34] Oftentimes, overcoming the embodied resource footprint can be challenging, particularly for material syntheses requiring high temperature processing and finite metal precursors.[35–37] In prior research, we examined cradle-through-use trade-offs between the footprint of carbon nanotube synthesis and the benefits that emerge from their product incorporation, finding clear net energy benefits of their use in certain electronic applications.[38] We also considered cradle-through-use trade-offs of nano silver-enabled textiles, where achieving benefits in the context of textile laundering were less achievable given the concentrations of silver in the textile.[34] Further research is necessary to understand the full suite of potential environmental benefits of material efficiency strategies utilizing nanomaterials.

In addition, once engineered nanomaterials are incorporated into a material system (*e.g.*, in nano-enabled composite), low dissipation is desirable to (1) reduce losses of invested resources to make the engineered nanomaterial, (2) maintain material integrity and performance, and (3) limit any potential hazard induced from engineered nanomaterial release (note: one exception is the intended function of antimicrobial action, wherein dissipation of the engineered nanomaterial from a composite is required, such as with nano-silver or nano-copper oxidation). Research to date indicates low dissipation of various engineered nanomaterial compositions from different bulk composite matrices (*e.g.*, carbon nanotubes and silver[39,40]), ranging from <1% to 4% of initial loaded mass. Considering low demonstrated dissipative potential and a circular materials goal of direct reuse of the nano-enabled composite (*i.e.*, no recycling, limited to no reengineering), this translates to no or very low potential end-of-life associated exposure concerns.

Nano-enabled concrete, an illustrative example

The example that we will use here to demonstrate how engineered nanomaterials can impact the properties of bulk materials and, possibly, advance the tenets of the circular economy is their addition to concrete, a fundamental component of global infrastructure. Concrete is the most used manufactured material on earth with a 50% projected growth of current usage by 2050, and its production generates 8% of global carbon dioxide emissions.[41] Approximately 82% of concrete is recycled in the U.S. in some form.[42] It is the case that, at present, no meaningful distinction is made between non-nano and nano-enabled concrete during recycling, thus whatever mechanical advantages nano-enabled concrete may have as a structural material is not considered to be a factor in recycling. A primary application of recycled concrete is as aggregate to concrete mixes, and most applications limit the recycled concrete component to about 20%.[43] Interestingly, engineered nanomaterials are proposed as an additive to such concrete mixes to compensate for any performance decreases as a result.[44,45] While the production and use of concrete generally follows major economic trends, the overall market for recycled concrete is expected to grow at a compounded rate of 7% through 2028, driven largely by the global focus on using less energy intensive,[46] lower embodied carbon, and circular materials. Thus, any advances that reduce the amount of concrete used and lengthens its lifetime will have a substantial impact on reducing global carbon dioxide emissions.

Typical aggregate mixtures for concrete consist of coarse grain (1–4 cm) and fine grain (<1 cm) fractions.[47] Very fine grains are usually excluded due to their contribution to premature weathering. However, the addition of small amounts of alkaline materials in the 1000–10 000 nm range, such as waste coal fly ash, has demonstrated some success improving workability, strength, and durability of concrete arising from its pozzolanic properties.[48] There has been a surge of interest incorporating nanostructured materials in concrete mixtures. The most popular of these are nano-silica, nano-alumina, and nano-clays, at amounts ranging from 1–5%, and carbon nanotubes at much lower concentrations (0.01–0.1%).[49–52] Recent efforts are pursuing more exotic nanostructured materials such as metal–organic-frameworks (MOFs).[53] The picture that emerges is one of nanomaterials extending the size range of aggregates used in concrete to enhance its properties.

Although results vary, such nano-enabled concrete mixtures have been found to increase mechanical strength, workability, ductility, flexural strength, and durability while lowering permeability.[54] Microscopy studies reveal nanoparticles blocking pores within the concrete structure, thereby retarding the movement of water into the matrix.[55] Carbon nanotubes with high aspect ratios can bridge across microcracks retarding their spread.[56–58] Such observations are noted to be consistent with improved durability, however tests are usually conducted during standard curing times (28–91 days). Concrete with added nano-silica and nano-alumina are used in practice, however few long-term results of full-scale nano-enabled concrete applications have been reported. Improved performance measures of nano-enabled concrete are consistent with circularizing a large-scale material critical to the construction industry. The combination of high mechanical performance and greater durability could make possible the use of smaller amounts of concrete with a longer life span and with greater demand for reuse. However, full realization of this potential will likely require parallel advances in technical standardization, structural design, planning, financing, and adaptive policies.

To demonstrate the potential to gain functional performance at a lower environmental cost for this example of nano-enabled concrete, we considered the existing literature data on improved performance due to nanomaterial additions. Concrete characteristics and performance metrics include tensile strength, compressive strength, flexural strength, Young's modulus, ductility, porosity, and water absorption. Here, we consider compressive strength at 28 days, normal concrete, and two nanomaterial additives with very different embodied energies, carbon nanotubes and nano-silica to present relative added environmental impact for the functional gain in compressive strength. We compare the additive options based on carbon equivalents (kg CO_2 equivalents) to improve the performance of 1 kg concrete (Table 1). The range of values for each category were identified from the literature. While the determination of the relative carbon footprint for each alternative is approximate, the exercise provides order-of-magnitude comparisons to inform tractable paths for engineered nanomaterials to support circular bulk materials.

Notably, the additional nano-silica needed to achieve the noted range in performance improvement (10–30% increase in 28 day MPa) is associated with the lowest carbon footprint, by orders of magnitude. Carbon nanotubes, on the other hand, carry a substantial embodied energy from synthesis and purification and thus, despite their extremely low addition rates, the added carbon footprint (regardless of multi- or single-walled) is greater than the comparative concrete addition needed to achieve the same function. Thus, until the energy required for synthesis of carbon nanotubes is substantially reduced, their addition for functional performance gains in concrete (at least for compressive strength) does not make sense in the current, circular bulk materials context.

While not comprehensive of all potential nanomaterial additives to concrete nor all metrics of performance improvements, this example illuminates important opportunities for small additions of certain engineered nanomaterials to have a substantial impact on lowering the carbon footprint of bulk composite materials by reducing the amount of material required and/or extending the material lifetime. The embodied resource footprint of synthesis is a critical factor in determining the potential benefits of a given nano-additive.

Summarizing thoughts

Recognizing the central role of materials in economic prosperity and societal well-being, the U.S. introduced the Materials Genome Initiative in 2011 to accelerate the discovery and deployment of materials.[63] By this time, engineered nanomaterials were established across research and development communities, having emerged in prominence after the 21st Century Nanotechnology Research and Development Act of 2003. The environmental nanotechnology community pursued research to evaluate risks and benefits of engineered nanomaterials in environmental and human matrices, from which conditions that define their safe manufacture, handling, and use emerged. Engineered nanomaterials are cornerstones of many technologies that elevate the U.S. global competitiveness in technology and defense, thus they are no longer emerging, they are fully integrated in society through the products that fuel our daily lives. As a result, these engineered nanomaterials are distributed throughout

Table 1 Concrete material additives and their associated carbon footprint to achieve a common increase in compressive strength

Added material	Increase % wt	% increase compressive strength (MPa, 28 d)	kg CO_2 eq. per kg material	C footprint[g] (kg CO_2 eq. for 1 kg concrete baseline)
Concrete[a]				
Cross sectional area[b] (columns, walls)	10–30%	10–30%	0.103	0.0103–0.0309
Volume (footings)[c]	7.5–22%	10–30%	0.103	0.0072–0.023
	% wt additive			
$nSiO_2$ [45,59,60]	1–5%	10–30%	0.0036[e]	0.000036–0.00018
CNTs[d] [61,62]	0.02–0.1%	11–23%		
SWCNT			180–6600[f]	0.36–6.6
MWCNT			60–2400[f]	0.012–2.4

[a] We determined the additional concrete mass that would be needed to achieve the same increase in compressive strength as that of the nano-additives. [b] $A = 1 - (1/1 + S)$, where A = % increase in cross sectional area, S = % increase in strength; based on ACI 318 equation 22.4.2.2 for maximum axial compressive strength. [c] $V = 1 - (1 + S)^{-3/4}$, where V = % increase in volume, S = % increase in strength; based on ACI 318 Table 22.6.5.2 for two-way shear strength assuming square column dimensions and depth proportional to the critical perimeter. [d] Most studies do not distinguish whether the CNT additive is multi- or single-walled or a mixture, so we include the range of minium to maximum values for both types. To convert embodied energy of CNT synthesis to kg CO_2 eq. per kg CNT, we use 74.9 kg CO_2 eq. per GJ of combusted heat, from the EcoInvent database version 3.9. [e] Value for fumed silica, a predominant method for producing nano-silica, extracted from EcoInvent database V3.9. [f] Embodied energy values from ESI† Table S1 in Gilbertson, *et al.*[32] [g] Using 1 kg amended concrete as a baseline comparison, this is the carbon footprint of the added material only: 1 kg concrete × % wt added × respective kg CO_2 eq. per kg material. For carbon nanotubes, the minimum and maximum values are indicated given the range in both wt% and embodied CO_2 eq.

environmental matrices, including in various forms of chemically and physically transformed from their original state. As adoption of the circular economy framework increases, we must consider the current and future horizons for revolutionizing materials in circular resource cycles, including with engineered nanomaterials.

There are many foreseen, and yet to be realized, opportunities for engineered nanomaterials to facilitate the shift in global material systems from linear to circular economy models of resource use. As presented herein, we envision an immediate opportunity is the use of certain engineered nanomaterials to improve the functional performance and/or durability of bulk materials. In many applications, the realization of these benefits is the primary technological reason that engineered nanomaterials are used in the first place. Yet there is little research on the natural resource savings of nanomaterial-enabled composite materials produced today or those being developed for future applications. We recommend that these indirect but potentially substantial material efficiency and circular economy benefits be quantified in nanoscience and engineering research. Further, quantifying these benefits using life cycle assessment or related tools could support the development of additional circularity metrics. The current predominant mass-based circularity metrics would miss material efficient advantages that engineered nanomaterials have to offer. As the incorporation of circular economy goals in sustainability certifications, standards, and government directives increases (*e.g.*, the European Green Deal as outlined by the European Commission[64]), there will be additional drivers to adopt such approaches.

Of the five circular economy business models outlined by the OECD,[7] engineered nanomaterial additives in composite materials are an example of product life extension. This model slows material cycles by extending product lifetimes to achieve resource efficiency. Here, we highlight how this could be envisioned for nano-enabled concrete, with substantial benefits in terms of environmental impact savings through the reduction of bulk concrete usage (*e.g.*, reduced amount of Portland cement, reducing repair/maintenance, increasing durability extending concrete useful life and/or facilitating reuse). We envision the next frontier for circular materials to be fueled by advances in material design and methods to computationally inform *a priori* design of nano-enabled composites. Machine learning and artificial intelligence methods are already being applied to the design of materials such as metal organic frameworks[65] and polymer nanocomposites.[66] Such approaches highlight the immense value in reducing the vast potential material design space to compositions that meet predefined physicochemical property criteria (*e.g.*, a minimum tensile strength or electrical conductivity). There remains an unrealized and tangible opportunity to establish and incorporate additional criteria that represent environmental benefits and impacts, material hazards, and circular material properties. Computational-aided sustainable material design would not only uncover currently undiscovered nano-enabled composite alternatives but would also expedite the use of engineering nanomaterials to improve resource efficiency and a circular economy and close our finite material loops.

Conflicts of interest

There are no conflicts to declare.

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