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Towards sustainable quantum dots: Regulatory framework, toxicity and emerging strategies

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ABSTRACT

Quantum dots (QDs) have garnered significant interest due to their unique optical and electronic properties, but concerns about their environmental and health impacts persist, particularly for those containing heavy metals like cadmium. The regulation of nanomaterials, including QDs, is relatively young, evolving, and often confusing. This review aims to provide a clear overview of the European regulatory framework, focusing on horizontal and sector-specific regulations that govern nanomaterials and QDs. These policies aim to manage the risks posed by these materials, ensuring safety across various industries. The toxicity mechanisms of QDs is then addressed, shedding light on the ways these materials can harm biological systems. In response to these challenges, sustainable approaches to classic QDs synthesis are critically examined, emphasizing safer production methods and reduced use of toxic reagents. Alternative to classic QDs, such as indium phosphide QDs and carbon dots, are presented as promising options to avoid toxic heavy metals. Sustainable synthetic routes for these alternatives are also discussed, highlighting the importance of selecting environmentally benign starting materials and utilizing continuous flow techniques. Together, these insights pave the way toward more sustainable and less toxic quantum dot technologies.

1. Introduction

Quantum Dots (QDs) are an attractive class of nanomaterials (NMs) that have earned significant attention from the scientific community since their discovery in the late 80 s, [1,2] culminating in the Nobel Prize in 2023. They are semiconductor nanocrystals with size smaller than the exciton Bohr radius, generally between 2 and 10 nm, that leads to quantum confinement effect. Due to quantum confinement, QDs exhibit unique properties lying between bulk materials and discrete atoms [3] and their energy band gap increases as particle size decreases, allowing their optical properties, such as quantum yield (QY) and redox potential, to be easily tuned. [4,5] The particle size is closely related to the synthetic approach used, as well as the selection of precursors and ligands, emphasizing the need for precise control over reaction parameters. Nowadays, QDs are synthesized from a variety of materials, like metal chalcogenides, [6] metal oxides, [7] metal halides, [8] and non-metal semiconductors. [9] Classically, the structure of QDs consists of a semiconductor core surrounded by a shell of organic ligands, which provide colloidal stability and facilitate charge mobility. However, a variety of QDs structures have emerged during the last years, such as alloys, different core-shell designs, and doped dots. [10] These differences result in diverse properties and behaviours, leading to a wide array of existing QDs. Among the unique properties of QDs, it is important to underline their up-conversion ability, tunable chemical activity, excellent photostability, large surface area and easy surface modification. [11,12] Thank to these characteristics, QDs became a prime subject of interest in a variety of fields such as photocatalysis, [13,14] drug delivery and biological applications, [15–17] optoelectronics and energy conversion and storage. [18,19] However, despite their promising features, the inherent toxicity of certain QDs, particularly those based on cadmium and lead, severely limits their applicability. [20] Various strategies have been developed to minimize this toxicity, such as the passivation of the core to prevent metal leaching or surface modification with biomolecules. [21] Although these modifications have made

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conventional QDs more promising, the development of safer and more environmentally friendly materials remains crucial. In particular, great efforts are being made for the development of indium phosphide (InP) QDs or carbon-based QDs (CDs), which are reported to be less toxic and more sustainable compared to traditional QDs. [22–25] Nevertheless, the luminescence of InP and CDs is still insufficient and further improvements are needed in order to use them in specific applications.

Evidently, while QDs offer many advantages, they also present notable challenges. The complex synthetic protocols, the use of hazardous reagents and the intrinsic toxicity of QDs hinder the use of these materials especially for industrial applications. Moreover, the advancements in the field of QDs, and NMs in general, confers a great impact with economic relevance in different areas of industry and research, making imperative to formulate regulations that govern their production and use. Although specific legislation in the European Union (EU) addresses NMs, the regulatory framework is still far from satisfactory. The struggle to harmonize NMs regulation and the drive for increased transparency regarding information on NMs span back more than a decade and is still ongoing. [26]

This review emphasizes the critical role of European regulations, directives, and restrictions concerning NMs and QDs. Legislation plays a pivotal role in shaping the production and use of QDs, with significant implications for both industry and research. Despite the extensive research on QDs synthesis, properties, and applications, regulatory guidelines remain underexplored. Understanding and aligning with these regulations is essential to ensure the safe development and commercialization of QDs, particularly in light of their associated toxicity. By linking regulatory frameworks with QDs toxicity, this review aims to provide the scientific community with valuable insights into how legal standards can support more responsible and effective advancements in the field. For this reason, Section 2 will offer an in-depth analysis of European laws governing nanomaterials, QDs, and their chemical constituents. Section 3 will focus on QDs toxicity mechanisms and the factors that influence them. Finally, Section 4 will explore sustainable synthesis of classic QDs and safer and more environmentally friendly alternatives, such as InP and CDs. The structure of the present review is depicted in Fig. 1.

The abbreviations used through the review are listed in Table 1 in alphabetical order.

Table 1
Abbreviations and definitions used in this review.

Abbreviation	Definition
AI	Artificial Intelligence
BDE	Bond Dissociation Energy
BPR	Back Pressure Regulator
CDs	Carbon Dots
CF	Continuous Flow
CHFS	Continuous Hydrothermal Flow Synthesis
CLP	Classification Labelling and Packaging
CNDs	Carbon Nanodots
CQDs	Carbon Quantum Dots
EC	European Community
ECHA	European Chemical Agency
EU	European Union
EUON	European Union Observatory on Nanomaterials
FWHM	Full Width at Half Maximum
GQDs	Graphene Quantum Dots
InP	Indium Phosphide
LHP	Lead Halide Perovskite
ML	Machine Learning
NMs	Nanomaterials
OA	Oleylamine
PTMS	Tris(trimethylsilyl)phosphine
QDs	Quantum Dots
QY	Quantum Yield
REACH	Registration, Evaluation, Authorization and Restriction of Chemicals
RoHS	Restriction of Hazardous Substances in Electrical and Electronic Equipment Directive
ROS	Reactive Oxygen Species
SVHC	Substance of Very High Concern
TOP	Trioctylphosphine
TOPO	Trioctylphosphine oxide
WEEE	Waste Electrical and Electronic Equipment Directive

2. Quantum dots in European legislation

As stated in the introduction, the fast development of NMs makes necessary an adequate regulatory framework and adapted regulations that consider the particularities related to nano- and quantum-technologies. During the last few years, legal acts have been written and revised worldwide in order to include rules on the use, production and labelling of NMs. However, there are significant differences among the various countries and in some cases, there are not even points of comparison regarding the aspects to be regulated or the standard type. [27] The United States (US) and the European Union (EU) are the main trailblazers in the establishment of regulations that contribute to the proper governance of nanotechnology; this review will focus mainly on the EU legislation. QDs are not directly addressed in the EU legislation but they have to respect regulations and directives in different fields. As depicted in Fig. 2, in order to understand how to handle QDs in EU legislation, it is possible to consider them regarding their size, their chemical composition or their use. Concerning their size, QDs are NMs and so they have to follow the regulatory system that concerns NMs. Another approach consists in considering the QDs for their chemical composition: several compounds have specific laws to regulate their production, import and/or use. Lastly, it is possible to find regulations that address chemicals and materials for their specific use: for example, rules must be followed for compounds that are considered “dual-use items”. Section 2.1 explains the general directives for the regulation of NMs and of specific chemicals (“size” and “composition”), while in Section 2.2 the regulation about dual-use items (“use”) will be discussed.

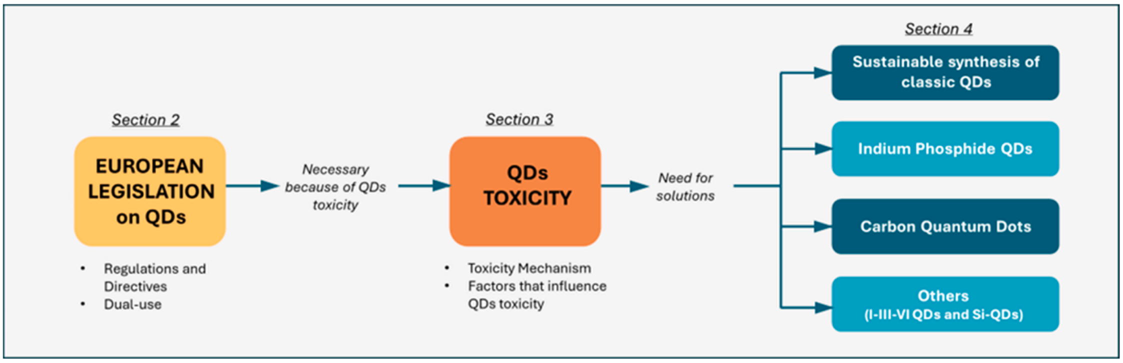


Fig. 1. Internal structure of this review.

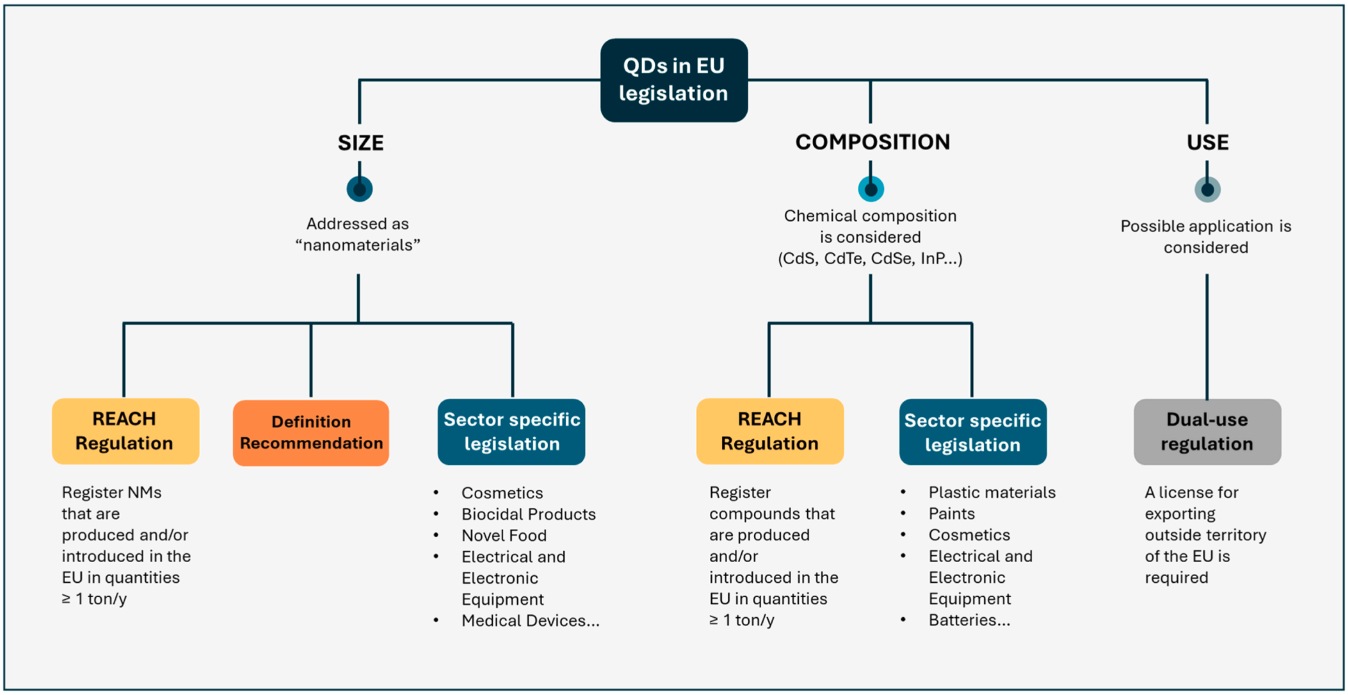


Fig. 2. Contextualization of QDs in the European regulatory framework. REACH = Registration, Evaluation, Authorization and restriction of Chemicals.

2.1. Regulations and directives

Before delving into NMs and QDs legislation, it is worth knowing that EU regulatory framework on chemicals can be divided into horizontal and sector specific legislations. The horizontal legislation (e.g., REACH = Registration, Evaluation, Authorization and restriction of Chemicals and CLP = Classification Labelling and Packaging) addresses issues broadly, while sector specific legislations consider specific problems for well-defined applications (e.g., cosmetic products, biocidal products, and medical devices). The European regulation that controls chemicals in general is the REACH regulation, the first version of which was adopted in 2006 and did not consider or make specific provisions for NMs. [28] Only after the genesis of different specific regulations and the introduction of a recommendation on the definition of a nanomaterial (2011), [29] the need to address nanotechnologies became impelling (see Fig. 3). In 2018 nanomaterials, or "nanoforms" as they are referred in the law, became explicitly considered in REACH where technical Annexes I (chemical safety assessment), III and VI-XII (registration information requirements) were modified with clarifications and provisions. [30] In 2020 the European Commission (EC) also updated Annex II, introducing new provisions on safety data sheets to complement the new registration requirements for NMs. [31] The recommendation on the definition of a nanomaterial of 2011 was reviewed and revised in 2022 leading to the actual definition adopted by the EU. [32]

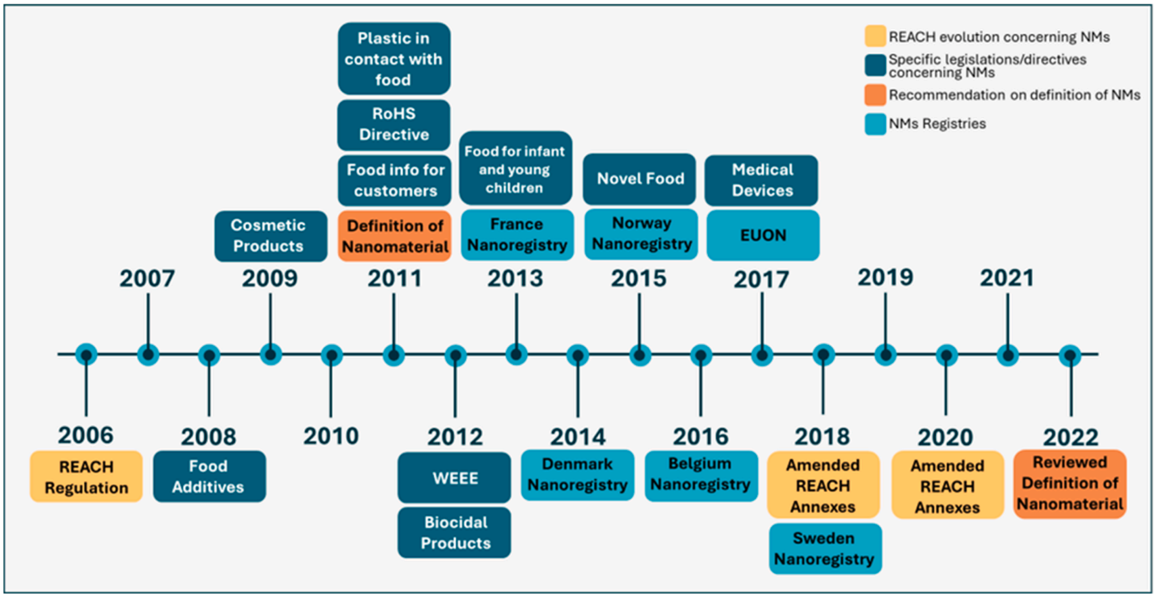


Fig. 3. Timeline of nanomaterial-relevant EU regulations and national nano-registries. REACH = Registration, Evaluation, Authorization and restriction of Chemicals; RoHS = Restriction of Hazardous Substances in Electrical and Electronic Equipment Directive; WEEE = Waste Electrical and Electronic Equipment Directive; EUON = EU Observatory for Nanomaterials.

The body of the law says: "'*Nanomaterial' means a natural, incidental or manufactured material consisting of solid particles that are present, either on their own or as identifiable constituent particles in aggregates or agglomerates, and where 50 % or more of these particles in the number-based size distribution fulfil at least one of the following conditions: (a) one or more external dimensions of the particle are in the size range 1 nm to 100 nm; (b) the particle has an elongated shape, such as a rod, fibre or tube, where two external dimensions are smaller than 1 nm and the other dimension is larger than 100 nm; (c) the particle has a plate-like shape, where one external dimension is smaller than 1 nm and the other dimensions are larger than 100 nm.*" This definition considers the only feature that all NMs have in common, i.e., their size. However, an implemented version is needed to take in consideration also the origin of the materials, the surface or the internal structure and products that contains NMs. Different European agencies have been formed with the aim of assisting the Member States and the European Commission with the implementation of the law through the publication of guidelines. ECHA (European Chemical Agency) is the executive agency related to REACH regulation and it can itself set bans, limits and conditions for manufacturing, placing on market and use of a chemical substance.

The current legislation under REACH expects companies to register chemical substances that are produced or imported in the EU in quantities equal to or more than one ton per year. The chemicals that are on the market below this threshold, but which are classified as hazardous from the CLP regulation, are however recorded in the EC-inventory of ECHA. [33] For the registration of a nanomaterial, together with the chemical safety assessment, information is to be provided on the production volume, use and safe handling, as well as on particle size, shape and surface properties of the nanoforms. [26]

In addition to REACH and CLP horizontal legislations, NMs are specifically addressed in different specific regulations or directives as represented in Fig. 3 in the dark green squares. It must be noted that the definition of NMs can differ from one regulation to the other. Some of these laws already mentioned "nano" before the EC recommendation of 2011 while others take in consideration the recommendation but differ in some level of detail. Table 2 reports some examples of the sector-specific regulations that comprise NMs, their definition and the way that NMs are addressed in the body of the law.

In addition to the examples reported in Table 2, other specific regulations that address NMs are: the regulation on plastic food contact materials, [34] on food additives, [35] on food intended for infants and young children or special purposes [36] and on the provision of food information to costumers. [37]

In order to gives information to the general public about NMs, ECHA created EUON (EU Observatory for Nanomaterials) [38] that collects information on NMs existing on the EU market. In addition to EUON, some EU member states have national NMs registry (e.g. Belgium, France, Denmark, Norway and Sweden – see light blue squares in Fig. 3). [26] In the reported horizontal and sector-specific regulations on NMs, QDs are never directly mentioned but they must be considered and treated as such. Moreover, different laws report limits and conditions for the most common substances that compose classic QDs: regulations discussing Cd and its compounds, CdSe, CdTe, CdS and InP are herein briefly discussed. Cadmium, and its compounds in general, is registered under the REACH regulation and is manufactured in and/or imported to the European Economic Area, at ≥ 1000 to < 10000 ton/year. It is considered fatal if inhaled and very toxic to aquatic life. Moreover, it may cause cancer, causes damage to organs through prolonged or repeated exposure and it is suspected to cause genetic defects, and to damage fertility or the unborn child. CdTe, CdSe and CdS are all registered under REACH and they are all considered harmful, if not toxic. [39–41] Moreover, ECHA listed CdS as Substances of Very High Concern (SVHC) because of its carcinogenicity and because of its properties causing adverse effects on kidney and bone tissues, making necessary further authorizations from the European Commission before use. [42] Annex XVII of REACH that regulates the restriction on the manufacture,

Table 2

Some sector-specific EU regulations or directives which consider NMs. The name of the regulation/directive, the definition of nanomaterial and the rules about them are reported.

Regulation or Directive	Definition of NMs	Law
Regulation on cosmetic products. [54]	An insoluble or biopersistant and intentionally manufactured material with one or more external dimensions, or an internal structure, on the scale from 1 to 100 nm.	The EC is to be notified of the content of NMs in cosmetic products together with their identification and foreseeable exposure conditions.
Regulation on the market and use of biocidal products. [55]	Active and non-active substances with the following characteristics: - 50 % or more of the particles have a size of 1–100 nanometers in at least one dimension; - Particles are in an unbound state or as an aggregate or agglomerate.	The approval of the active substance does not cover the nanoform of the active substance except where explicitly mentioned. A separate dossier with all data requirements must usually be prepared for nanoforms of active substances. A dedicated risk assessment is needed.
Regulation on novel food. [56]	Any intentionally produced material that has one or more dimensions of the order of 100 nm or less or that is composed of discrete functional parts, either internally or at the surface, many of which have one or more dimensions of the order of 100 nm or less, including structures, agglomerates or aggregates, which may have a size above the order of 100 nm but retain properties that are characteristic of the nanoscale.	Food consisting of engineered NMs should also be considered a novel food and thus be subjected to all the regulation.
Regulation on medical devices. [57]	A natural, incidental or manufactured material containing particles in an unbound state or as an aggregate or as an agglomerate and where, for 50 % or more of the particles in the number size distribution, one or more external dimensions is in the size range 1–100 nm; Fullerenes, graphene flakes and single-wall carbon nanotubes with one or more external dimensions below 1 nm shall also be deemed to be nanomaterials.	Devices shall be designed and manufactured in such a way as to reduce as far as possible the risks linked to the size and the properties of particles which are or can be released into the patient's or user's body, unless they come into contact with intact skin only. Special attention shall be given to NMs. All devices containing NMs are classified based on the potential internal exposure of the patient.
Directive on Waste Electrical and Electronic Equipment (WEEE) [58] and on Restriction of Hazardous Substances in Electrical and Electronic Equipment (RoHS). [46]	No specific definition is given.	NMs for which specific treatment may be necessary are mentioned, but no specific requirements or definitions have been introduced.

placing on the market and use of certain dangerous substances, mixtures and articles, put limits on the amount of Cd and Cd derivatives in different products. [43] For example, the concentration of Cd in mixtures and articles produced from a list of plastic materials (e.g. polyurethanes, polyvinylchloride, epoxy resins etc.) cannot exceed 0.01 % by weight of the plastic material. The same threshold at 0.01 % by weight of Cd is set also for some type of paints, for brazing fillers and for jewellery among others. In addition, cadmium and cadmium compounds are banned or restricted under several specific regulations. The Annex II of the regulation on cosmetic products, for instance, lists Cd and its compounds among the banned substances from use in any cosmetic product for sale or use in the EU. [44] Also the regulation on batteries of 2023 states that Cd should not be contained into batteries. [45] This regulation, however, does not apply if the batteries are for equipment connected with the protection of Member States' essential security interests, arms, munitions and war material and for equipment designed to be sent into space. Portable batteries shall not contain more than 0.002 % of cadmium (expressed as cadmium metal) by weight. The RoHS (Restriction of Hazardous Substances in Electrical and Electronic Equipment) Directive (Annex II) restricts Cd compounds to 0.01 % by weight to any homogeneous material in a product. [46] This means that each smaller homogeneous part of a product (e.g., each LED in a screen) cannot have more than 0.01 % of Cd by weight. An exemption to this limit (starting in January 2025 and finishing in December 2027) allows the use of 5 $\mu g/mm^2$ for QDs directly deposited on LED semiconductor chips for use in display and projection applications. [47] Other products that are not supposed to follow the restrictions set by the regulation, are, for instance, equipment for military or security purposes, equipment to be sent into space, large-scale stationary industrial tools, R&D equipment and photovoltaic panels among others.

Considering the toxicity of Cd compounds (see Section 3 for further details) and the strict regulations in regards of their use, the need for safer alternatives becomes compelling. InP QDs are considered as the most promising candidate to replace Cd-based QDs thanks also to the low toxicity claimed in several scientific studies (see Section 4.2). [48–50] InP is registered under the REACH regulation being manufactured in and/or imported to the European Economic Area, at ≥ 1 to < 10 ton/year. This semiconductor is suspected to be carcinogenic and to damage fertility [51] and, very much alike Cd-based compounds, its presence is banned in cosmetic products under the Cosmetic Products regulation (Annex II). [52] Being classified as carcinogen category 1B (i.e., presumed to have carcinogenicity), the restrictions for substances under REACH Annex XVII prohibit the supply of InP to the general public as a substance, as mixtures or as a constituent of other mixtures. However, the use of the substance in electrical and electronic equipment would generally not be considered as a supply of the substance to the general public (neither as a substance, as a mixture or as a constituent thereof) as reported in Annex II of RoHS directive “Dossier for Indium Phosphide”. [53] The same Annex states also that a restriction of InP is not expected to generate substantial benefits for the environment or for health, since potential alternatives (especially GaAs for optoelectronic devices and Cd-based QDs for displays/lighting) are not considered to have a better environmental performance than InP.

2.2. *Dual-use*

NMs and QDs remain at the forefront of scientific research, with advancements in these fields accelerating rapidly. However, additional attention must be given to the fabrication and export of these materials, as they can be classified as dual-use items. Dual-use items encompass technologies and software that can be applied for both civilian and military purposes, including materials and chemicals potentially used in the design, production, or deployment of nuclear, chemical, or biological weapons. The EU addresses dual-use items in Regulation (EU) 2021/821 (the "Dual-Use Regulation"), [59] which revises previous export control rules. Annex I of the regulation reports an exhaustive list of all the goods that have to be considered and treated like potentially useful for military applications; this annex is dynamic to account for technological advances over time and it is updated every year. All items that are listed require a license for exporting outside territory of the EU. Each dual-use item is assigned a classification code, which combines numbers and letters to indicate the item's category, subcategory, and specific control entry. These classification codes are essential for the authorization and export documentation processes, referring to both the nature of the item and the control regime that governs it. An example classification of hetero-structured semiconductor electronic devices is shown in Fig. 4.

The EU's dual-use regulations do not specifically address QDs, but several classifications encompass them. For instance:

- Classification 3E003 includes hetero-structured semiconductor electronic devices (e.g., quantum wells, superlattices).
- Classification 6C004b covers some electro-optic materials like $AgGaSe_2$ or GaSe.
- Classification 6A004b comprises optical components made from ZnSe or ZnS with specific characteristics.

As can be seen, these classifications consider not only the chemical composition and physical state of materials but also their potential applications. Indeed, researchers and academics should be aware of some research scenarios that may trigger export controls, such as:

- Teaching, consulting, collaborating or working on research involving dual-use items with visiting foreign researchers inside the customs territory of the EU.
- Organizing or presenting at a conference/meeting/seminar about research involving dual-use items.
- Publishing about listed dual-use technology.
- Submitting information for patent application and patented information.
- Exporting tangible dual-use items (goods), including prototype design and lab equip ment.

To help academics understanding and complying with the Dual-Use Regulation, a recommendation has been published. [60] The recommendation is a non-legislative act that contains guidelines and tips to help managers and researchers understand and follow the current law. However, since the regulation's enactment in 2021, the global context for export controls for dual-use items has fundamentally changed due to geopolitical events. In January 2024, the European Commission released a white paper proposing EU actions in export control, sparked by recent geopolitical shifts. [61] A white paper is a document with a proposal for EU action in a specific area that is used to launch a debate with the public, stakeholders, the European Parliament and Council to arrive at a political consensus. On one hand, the Russo-Ukrainian conflict necessitates rapid restrictions on dual-use items to hamper Russia's military capabilities. Simultaneously, individual EU Member States have imposed new controls on emerging technologies, potentially fragmenting the single market and posing regulatory inconsistencies. For example, the Netherlands introduced tighter NM export restrictions in 2023, while Spain and Finland implemented stricter controls on quantum technologies. These developments raised questions about the adequacy of the current EU regulation to effectively contribute to the security of the Member States. Another recent example of stricter export control adopted autonomously from a Member State is the “Regulation on additional control measures to the Dual-Use Regulation” entered into force in the Netherlands in December 2024. [62] The Dutch government decided to implement the controls for export of certain emerging technologies deriving from the semiconductor, quantum and additive manufacturing sectors. The Annex of the Dutch Regulations lists goods, software and technologies that are not listed in Annex I to the EU Dual-Use Regulation 2021/821 and that need a further licensing

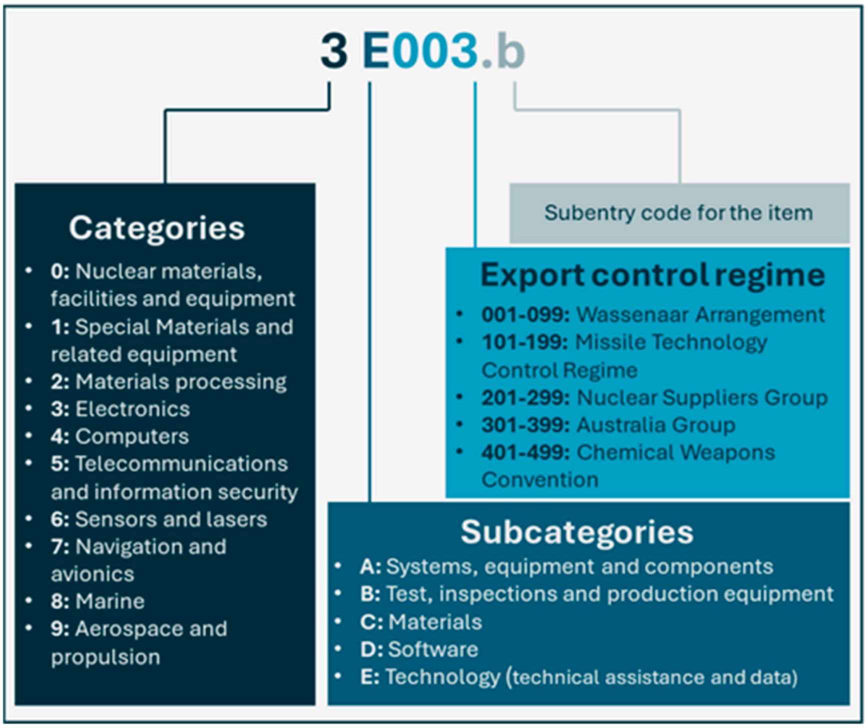


Fig. 4. Example of dual-use classification number for hetero-structured semiconductor electronic devices.

requirement for their export outside the Netherlands.

Understanding the legislative background is crucial for researchers in quantum- and nano-materials, as these areas are considered not only scientifically relevant but also strategically significant for military applications. [63] Nanotechnology is already widely used in military and research and development is expanding rapidly spanning potential future applications in all areas of warfare. [64] The World Health Organization has highlighted that NMs may pose both accidental and deliberate risks due to their neurotoxic and bioaccumulative properties, with direct implications for defensive and offensive military technologies. [65,66]

Concerning specifically QDs, several examples of applications for military uses can be found in the literature. Raytheon Company, a former major US defense contractor now merged into RTX Corporation, published different patents reporting military applications of QDs. For instance, they developed a countermeasure system to protect an asset from a missile attack composed by a decoy made with QDs that emit radiation with a profile similar to the asset. [67] Always Raytheon company published another patent on ‘Quantum dot-based identification, location and marking’ for military and recovery missions. [68,69] Lockheed Martin, one of the biggest US defense contractors, published as a patent the use of entangled quantum particles for the fabrication of radar systems. [70] Additionally, $AgInS_2$ QDs have been found effective for detecting trinitrotoluene at trace levels (6 μM), as demonstrated by research from the Chemistry Branch of the US Naval Air Warfare Center. [71]

The potential of QDs in quantum computing further underscores their dual-use implications. Since the concept of the spin qubit quantum computer was proposed in 1998 there has been a growing interest in using nanocrystals for quantum technology applications (see Fig. 5). [72] The spin qubit quantum computer is a quantum computer based on the control of the spin charge of carriers in QDs. While early systems utilized GaAs-based QDs, [73] significant advances have been made with silicon [74,75] and graphene-based qubits. [76] In order to accelerate the development of quantum technologies, the US Naval Research Laboratory, published a paper reporting the possibility to tune multiple QDs into resonance within the same waveguide and demonstrate a quantum interaction via superradiant emission from three QDs. [77]

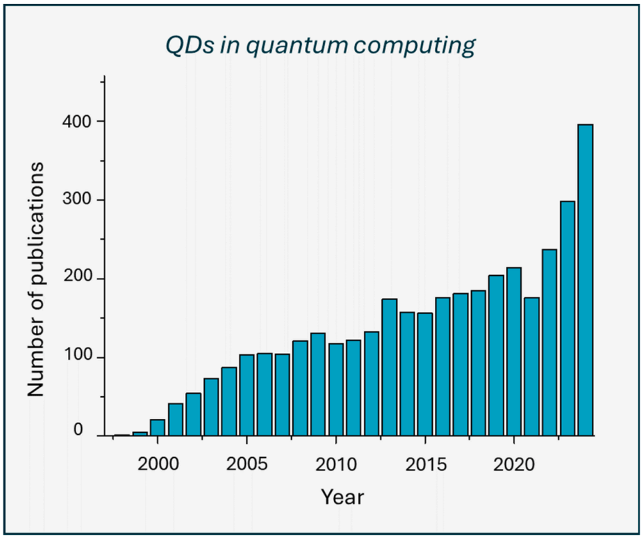


Fig. 5. Number of publications per year regarding the use of QDs in quantum computing. Data from Scopus using the keywords “qubits” and “quantum dots”.

The use of QDs in quantum computing would open a wider debate since quantum technologies are considered as a big portion of dual-use items. Quantum technologies can be involved in several aspect of offensive and defensive military applications and their use in this field introduce new capabilities, improving effectiveness and increasing precision, leading to the so called “quantum warfare”. [78] Although commercial-scale QD-based quantum computing remains a future goal, ongoing research and development are essential for bringing this technology to fruition. [79]

3. Toxicity of quantum dots

The majority of the regulations discussed in Section 2 pertain to the environmental and human health hazards posed by NMs and QDs. Although interest in QDs continues to grow, their practical applications are often limited due to their intrinsic toxicity. To address this, strategies for reducing or minimizing QDs toxicity are actively being developed (see Section 4), alongside ongoing studies on their potential harm. [80] This section will delve into the toxicity of QDs, with a particular focus on the factors that influence it. However, the analysis of toxicity data remains challenging due to the wide variety of QDs, each with distinct physicochemical properties, and the complex nature of biological systems. [21,81]

3.1. Toxicity mechanisms

Before exploring the factors influencing QDs toxicity in detail, it is essential to understand the primary mechanisms underlying their cytotoxic effects. The cytotoxicity of QDs generally follows three main pathways, as illustrated in Fig. 6. [82,83] First, the core of conventional QDs can interact with biological media, leading to its degradation and the subsequent release of toxic heavy metal ions, such as Cd^{2+} [84] and Ag^{+}. [85] In addition, QDs can produce Reactive Oxygen Species (ROS, e.g., superoxide, hydroxyl radicals and singlet oxygen) which increase intracellular oxidative stress and induce apoptosis. [86] While ROS play critical roles in cellular processes, their levels are typically regulated by antioxidants, as excess ROS can cause irreversible damage to cells and tissues. [87] For example, cadmium is known to generate ROS by altering antioxidant enzymes, including catalase [88] and superoxide dismutase. [89] In general, QDs can cause ROS excess by reducing the antioxidant system [90] or by inhibiting complex II and III in mitochondria. [91,92] A third toxicity mechanism involves the non-specific adsorption of QDs onto the cell membrane. This interaction can lead to cell membrane damage, structural alterations in membrane proteins, and disruptions in the transport of substances across the membrane. [93,94]

3.2. Factors that influence quantum dots toxicity

The toxicity of QDs is influenced by intrinsic properties such as chemical composition, size, shape, charge, and surface modifications. Additional factors, including concentration, exposure route, and stability, also play a crucial role. These aspects are discussed in detail in this section.

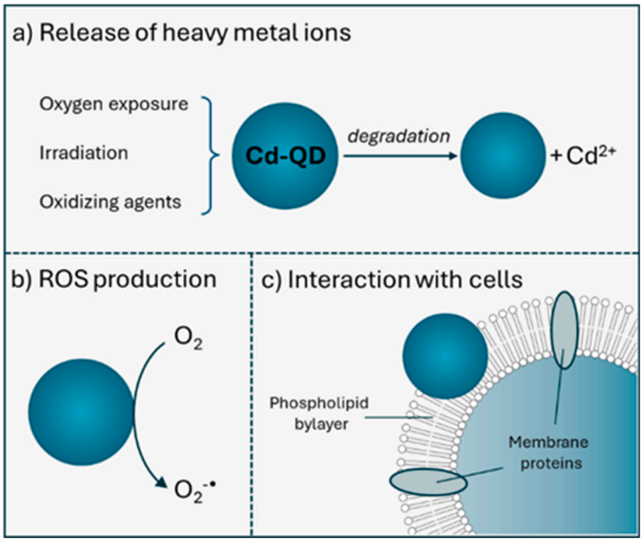


Fig. 6. Possible toxicity mechanisms of QDs: a) degradation of the heavy metal core and release of toxic ions (example for Cd-based QDs); b) production of reactive oxygen species with consequent induced apoptosis; c) adsorption on cell membrane and interaction with membrane proteins.

3.2.1. Chemical composition

The cytotoxicity of metallic QDs is largely determined by their core composition. The metallic core can release metal ions that interact with biological environments, with toxicity varying depending on the type of metal. The most studied and used QDs are Cd-based, such as CdS, CdTe and CdSe. These QDs exhibit bright and photobleaching-resistant emission together with other interesting properties that make them suitable for different applications, in particular in biomedicine. [95] However, cadmium is toxic and carcinogenic, posing health risks to humans and animals. Cd exposure, particularly to Cd^{2+} ions, has been linked to various cancers (breast, lung, prostate, nasopharyngeal, pancreatic, and kidney) and osteoporosis. [96–98] To mitigate the toxicity of Cd-based QDs, different types of coating or shells have been used. However, after contact with biological systems the coating can degrade exposing the Cd^{2+} core. [99] Other examples of composition toxicity can be found in Ag or Ti-based QDs also if the effects produced by these QDs result to be far way less serious than Cd. [100] Consequently, research has shifted towards Cd-free QDs, such as those based on indium phosphide (InP), [101] zinc selenide (ZnSe) [102] or copper indium sulfide ($CuInS_2$). [103]

3.2.2. Size

Particle size and shape have obvious effects on the toxicity of QDs. Generally, it is considered that smaller particles exhibit higher toxicity: [104] very small particles can be absorbed by the human body, enter rapidly the blood circulation and distribute into most of the organs. [81] An example was given by Maysinger and co-workers: two negatively charged CdTe QDs with diameter of 2.3 and 5.7 nm were synthesized and their toxicity on different cell lines was tested. At the same concentration, the smaller QDs were significantly more toxic than the bigger ones in all the tested cell lines. [105] This effect can be explained in two ways: (I) the smaller is the size the larger is the specific surface area and the more metallic core is exposed leading to more opportunities to form toxic metal ions; (II) many pathways in the body are size-dependent and allows only particles with a diameter below a certain threshold (e.g., the placental barrier or the nuclear pore complex). [100] Other studies, however, demonstrated different results. He et al., showed how smaller QDs are removed from the circulatory system faster than the larger ones that, in turn, lead to accumulation and severe toxicity. [106] Su et al. demonstrated how the biodistribution of QDs is size-dependent: QDs with larger sizes cumulates quickly in the spleen while smaller ones are more easily absorbed by the kidneys. [107]

3.2.3. Surface functionalization and charge

As previously mentioned, QDs are formed by a semiconductive metallic core and a possible passivating shell together with stabilizing ligands and functional groups on the surface. The nature of the shell, the ligands and the functional groups is an important parameter to evaluate in order to reduce the toxicity of QDs. The presence of a shell, usually made of another semiconductor material (e.g. ZnS), can help in decreasing the toxicity of the core preventing the leakage of heavy metal ions while improving the luminescence. Also, the presence of carefully selected ligands can help decreasing the toxicity of QDs. Liu et al., for instance, demonstrated how using a chitosan derivative (poly(ethylene glycol)-graft-chitosan) as a ligand for CdSe helps reducing the toxicity of the QDs. [108] The nature, and more importantly the charge, of the functional groups present on the surface plays also a crucial role in the assessment of QDs toxicity. The surface charge influences the rate of QDs transport through the plasma membrane, as well as their interaction with inner cell components. [109] Generally, it would be easy to affirm that, since the majority of biological membranes have a negative charge, the most toxic QDs are the one characterized by a positive charge that allows their easy absorption on the cell. [110,111] However, surface charges influence toxicity in a cell-specific way, since some cells are characterized by an increased sensitivity to positively charged QDs, [112] and others are more susceptive to the action of negatively charged ones. [113]

For example, Xiao et al. showed how negative and neutral QDs are preferentially distributed in the liver and spleen, while positive ones resulted to be mainly deposited in the kidneys confirming the crucial role of surface charge in QDs biological interactions. [112] Another study on the influence of the surface functionalization on the toxicity of QDs is given by Song et al. [114] In their work CdSe/ZnS have been modified with four different ligands (anionic poly(ethylene glycol)-carboxyl, anionic mercaptopropionic acid, zwitterionic glutathione, and cationic cysteamine) and tested for their in vivo toxicity on zebrafish. The results showed that surface charge plays a pivotal role in the adsorption and accumulation of the dots. Cationic cysteamine-QDs resulted in higher absorptions than the anionic and zwitterionic ones but anionic poly(ethylene glycol)-carboxyl-QDs induced the most severe toxicity. This suggested that the surface charge cannot be the sole factor influencing QD s toxicity, but all the parameters already listed (size, functionalization and composition) must be considered. All these parameters, however, have a different impact on the overall toxicity of QDs with the composition of the core playing the major role. In their publication, Iyer et al. ranked the contribution of charge, ligand length and size of the particle on the toxicity of QDs. [115] In particular, the impact of charge on toxicity is the most significant, with positively charged QDs being more toxic. This is followed by ligand length, where longer ligands result in higher toxicity than shorter ones, and finally by particle size, with smaller particles being more toxic.

3.2.4. Concentration (exposure time/dose)

QDs toxicity was confirmed to be directly dependent on their concentration and so on the exposure time and dose. [116,117] Several studies demonstrated the dose-dependent activity of QDs. Lau et al., for example, studied the chronic effects of CdTe QDs with three different sizes on human bronchial epithelial cells. [118] The two smaller dots resulted to be cytotoxic and carcinogenic in a dose-dependent manner while the larger QDs had negligible toxicity. Similarly, Roberts et al. demonstrated that QDs dose correlates with the severity of lung damage in studies involving CdSe/ZnS QDs with different functional groups (carboxyl or amine). [119]

Although an increasing number of studies is focusing on the cause of QDs toxicity, there is still limited understanding of the mechanisms, bioaccumulation, organ-specific effects and impacts on organisms in real exposure scenarios. [81] Additionally, the wide variety of QDs and biological environments complicates standardization of tests and methods leading to even more complicated data comparison. In general, the toxicity of QDs can be attributed to four key factors in decreasing order of impact: chemical composition, surface charge, ligand/shell nature, and particle size (Fig. 7). However, the overall effect results from the synergy between all these physicochemical properties and the biological environment that is exposed.

4. Exploring safer alternatives for quantum dots production

Over the past two decades, the most studied QDs have primarily been binary compounds made from elements in groups II–VI such as CdS, CdTe, CdSe and ZnS. However, the potential toxicity of heavy metals in these QDs presents significant challenges to their application. Additionally, their synthetic approaches require expensive equipment, stringent experimental conditions, toxic raw materials, and pose potential environmental risks, further limiting their commercial development. Consequently, developing environmentally friendly synthetic methods became a top priority for researchers, followed by the search for greener, non-toxic alternatives to traditional QDs. This section will discuss the main strategies for safer and more sustainable syntheses of classical QDs (mainly II-VI group) and two families of promising, less-toxic alternatives: InP QDs and CDs.

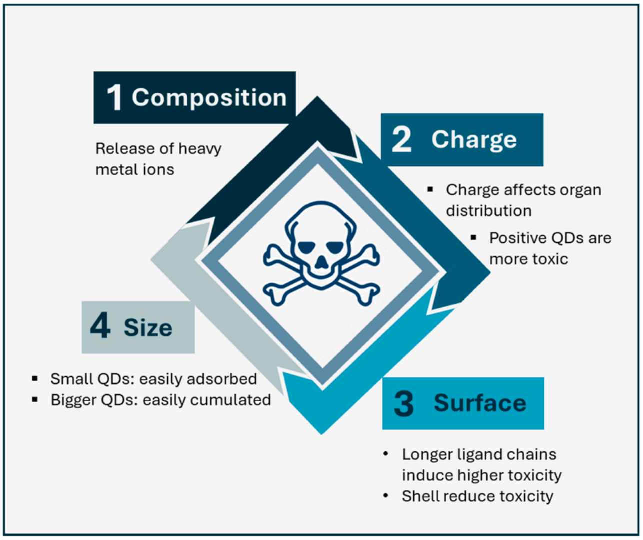


Fig. 7. Main factors affecting the toxicity of QDs.

4.1. Sustainable approaches for classic II-IV quantum dots synthesis

The primary methods for preparing classic QDs are colloidal chemical techniques including organic [120] and aqueous phase syntheses. [121] Organic synthesis is a traditional approach that involves high temperatures, oxygen-free atmosphere and the use of hazardous precursors, ligands and solvents, such as dimethyl cadmium ($CdMe_2$), trioctylphosphine oxide (TOPO) and trioctylphosphine (TOP). [122] Great efforts have been made to transform the synthetic pathway towards more sustainable options. Namely, a greener strategy for synthesizing QDs was introduced in a study by Peng and coworkers whom obtained quasi-monodisperse Cd chalcogenide QDs replacing $CdMe_2$ with CdO. [123] Moreover, this approach eliminates the need for strict anhydrous condition, significantly simplifying the process while reducing environmental pollution. CdS, CdSe and ZnSe QDs were also synthesized using oleic acid as a cheap and safe ligand (in opposition to TOP and TOPO) and octadecene as a non-coordinating solvent. Likewise, the group of Tang and Zou proposed a green, cost-effective method for CdSe QDs synthesis using paraffin liquid and oleic acid as the reaction medium. [124] ZnSe QDs have been synthesized from Jiang et al. using environmentally friendly *N*,*N*-dimethyloleoyl amide to dissolve Se powder at mild temperatures, with oleic acid acting as a natural surfactant to solubilize $Zn(OAc)_2$. [125]

In order to limit the use of hazardous organic solvents, aqueous-phase synthesis of QDs, involving the use of inorganic precursors with suitable stabilizers in water, started to expand. [121] This method offers several advantages, including simplicity, low cost, and the ability to control surface charge. Rabinal et al. reported an efficient green method to prepare mercaptopropionic acid-capped CdSe QDs at room temperature using a stable hydrazine hydrate-Se complex. [126] The resulting CdSe QDs exhibited adjustable particle sizes ranging from 1.58 to 3.42 nm by varying the annealing time, as confirmed by band-edge absorption analysis. Recently, different type of capping agents for the aqueous preparation of QDs have been investigated in order to achieve greener and more biocompatible materials. Among the variety of the stabilizers that can be found in the literature, it is worth noticing the advent of more natural sources such as amino acids [127] or plant extracts. [128,129]

Recently, Monbaliu et al. reported the use of tris(2-carboxyethyl) phosphine as an efficient and water soluble vehicle for the chalcogenide transfer for the synthesis of CdS, CdSe, CdTe and CdSe/ZnS. [6] Moreover, the aqueous preparation of these QDs was successfully intensified both in a microfluidic and a mesofluidic setup through superheated flow conditions. [130] Continuous flow chemistry is, indeed, recognized as one of the most promising tools to overcome the limitations of classical batch synthesis of NMs and obtain tailored products in a more controllable and reproducible way. [131,132] Along with precise control over various reaction parameters, continuous flow technology allows a safer handling of toxic and hazardous materials and leads to a more sustainable production of QDs. [133,134] A significant example is reported in the work by Jensen et al. whom fabricated a stainless-steel tubular microreactor for the formation of ZnSe QDs. [135] A gas–liquid flow was adopted in the microreactor to eliminate parabolic residence time distribution, resulting in narrow particle size distribution.

Another promising approach to enhance the sustainability of QDs synthesis is through accelerated discovery using self-driving labs, artificial intelligence (AI), and machine learning (ML). Modular, reconfigurable flow synthesis platforms, which incorporate precursor formulation, controlled flow synthesis, and in situ diagnostics, allow high-throughput experimentation with real-time analysis of QDs properties. [136] Abolhasani and coworkers have conducted extensive research on automated synthesis for various types of QDs, including Cd-based and lead halide perovskite QDs (LHP QDs). In particular, their work leverages autonomous flow reactors to create a streamlined and reproducible process, using advanced AI-driven setups to refine synthetic parameters in real time. This automation facilitates the production of high-quality QDs with optimized properties. Their work includes research on closed-loop systems, such as the "Artificial Chemist" designed to accelerate material discovery and parameter optimization by autonomously adjusting synthesis conditions based on real time optical measurements and predictive models. [137] An automated microfluidic platform has been used, for instance, to study the continuous biphasic synthesis and ligand exchange process of CdSe QDs. [138] In particular, the platform was designed to improve control over biphasic segmented flow, mitigating issues that often arise in CF systems due to the adherence of nanocrystals to microchannel walls. In another study, Abolhasani's group presented AlphaFlow, a reinforcement learning-guided self-driven laboratory system designed with modular fluidic processing units for autonomous synthesis exploration. AlphaFlow focused on complex multi-step reactions which are used to create precise hetero-nanostructures. Using CdSe/CdS core-shell QDs as a test case, AlphaFlow autonomously explored multi-step reaction pathways and independently identified optimal reagent sequences, even without conventional knowledge of chemical order, resulting in QDs with improved absorption and quality. [139] An autonomous, modular microfluidic system with AI-driven decision-making to optimize LHP QDs synthesis has also been developed. [140] This system controls 10 parameters, enabling rapid optimization for 10 QDs types with distinct emissions. Additionally, the so called "Smart Dope" platform accelerated the CF synthesis of multi-cation-doped LHP QDs, combining digital twin modelling with closed-loop optimization to streamline high-performance QDs production and discovery of new synthesis routes on demand. [141]

Mechanochemical methods are also being developed for the preparation of Cd-based QDs with the aim of avoiding the use of hazardous solvents. Shalabayev et al., for instance, reported the mechanochemical synthesis of CdS using cadmium acetate and sodium sulfide. [142] Other alternative green methods to obtain QDs are the ones based on the use of biological organisms (bacteria, viruses, algae and fungi among others). The biosynthetic processes lead to QDs with inherent biostability and biocompatibility and are very specific avoiding the formation of any by-product. [143] An example is given in the work from Liu et al. were CdSe QDs were prepared in vivo using the yeast *Candida utilis*. [144] Interestingly, highly fluorescent and photostable QDs were obtained and used directly for bioimaging applications. Another strategy to limit the hazardousness of Cd-based QDs is based on the post-reaction modification of the nanocrystals. Passivation of the metallic core with polymeric or semiconducting shell coatings prevent the leaching of metal ions and improve the stability of the materials. Moreover, surface modification with water soluble ligands or biomolecules provide bioconjugation approaches for toxic QDs. [21]

In summary, to achieve a more sustainable synthesis of classic II-IV QDs different strategies can be adopted as depicted in Fig. 8. Firstly, toxic and hazardous precursors can be substituted with greener and safer ones. Moreover, different synthetic approaches can be used: lowering the temperature, using more sustainable solvents, using a continuous flow setup and self-driving labs or developing alternative synthesis like biosynthesis or mechanochemistry. Lastly, QDs can undergo a post-synthetic modification to grow a protective shell or to passivate the surface avoiding metal leaching.

To reduce the environmental impact of Cd-based QDs, recycling their metal components is essential. Common techniques involve acid leaching, using nitric acid [145,146] or a mixture of sulfuric acid and hydrogen peroxide. [147,148] For example, Fthenakis et al. used a dilute sulfuric acid and hydrogen peroxide solution to leach Cd and Te from CdTe-based photovoltaic modules, achieving then complete separation through cation-exchange resins. [148] To avoid the generation of wastewater or waste gas during the recycling process, Liu et al. proposed a vulcanization-vacuum distillation method to retrieve Te from CdTe, achieving 99.92 % pure Te. [149] This process offers advantages such as no waste gas, efficient resource use, a short refining process, and compliance with energy-saving and clean production standards.

Despite efforts to improve the synthesis and reduce the toxicity of Cd-based QDs, concerns regarding the environmental and health risks associated with heavy metals remain unresolved. As a result, the development and use of less toxic QDs has become an important area of research. Two primary alternatives have garnered significant attention from the scientific community: heavy metal-free QDs and metal-free QDs. In the following sections, one example from each category will be explored: namely, indium phosphide (InP) QDs and carbon dots (CDs). Their properties, along with sustainable synthetic methods, will also be discussed.

4.2. *Indium phosphide quantum dots*

As an environmentally benign material belonging to the III–V group semiconductor, InP is deemed as another promising candidate to replace Cd-based QDs. [150] Several studies, indeed, claim the safer nature and

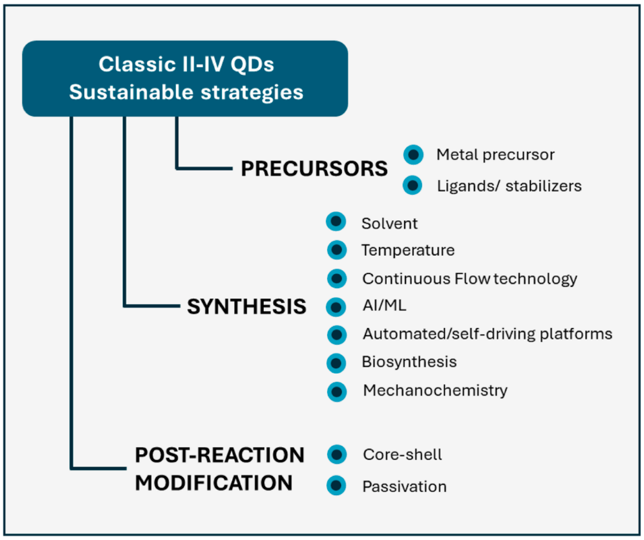


Fig. 8. Main strategies to achieve a greener synthesis of classic QDs.

decreased hazardousness of InP when compared to classic II-IV nanocrystals. [23,151,152] For example, Pompa et al. demonstrated the lower toxicity of In-based materials confronting CdSe/ZnS and InP/ZnS both *in vivo* and *in vitro.* [22] Selecting QDs with similar physical and chemical properties, they observed almost identical cellular uptake and metal leaching (Cd^{2+} and In^{3+}). However, InP/ZnS resulted to have very low toxicity in opposition to CdSe/ZnS showing that In-based QDs can be a safer alternative to Cd-based ones.

Another feature that makes InP QDs an interesting alternative is their bandgap of 1.35 eV that allows a broad wavelength of emission by varying the nanocrystal size, potentially covering the whole visible range from blue to deep red. Progress in replacing Cd-based QDs with InP QDs has, however, faced challenges. InP QDs, with their higher covalency, are prone to forming deep trap states that capture excitons. Moreover, the increased surface-to-volume ratio at smaller sizes makes surface conditions critical to performance and the presence of uncoordinated dangling bonds and lattice disorders in surface chemistry can lead to a depletion of InP optical properties. These surface defects, indeed, introduce new surface-state energy levels disrupting the band structure and acting as trap states. This results in emission quenching, reduced photostability, and deviations from expected luminous efficiency and narrow emission linewidths. [153]

The preparation of high-quality InP has, indeed, proven challenging compared to type II-VI QDs, likely due to their highly covalent nature. However, thanks to continuous efforts, the fluorescent properties of InP QDs have been rapidly improving, approaching the performance of their II-VI counterparts. Most synthetic methods for InP QDs involve reacting an indium source (commonly an indium salt or fatty acid) with a phosphorus source in a high-boiling coordinating or non-coordinating solvent at elevated temperatures. Although as-synthesized core InP QDs generally show lower photoluminescence QY, their core/shell structures, usually coated with a ZnS shell, demonstrate high efficiency. [154–156] The passivation of InP surface has proven to be an efficient tool to increase the stability and the photoluminescence QY. Tang et al., for example, recently post-treated InP using cysteamine effectively passivating surface defects and non-radiative sites, enhancing the stability and reaching a lifetime of over 1200 h. [157] Also zinc myristate have been successfully used with this scope: thanks to an *in-situ* passivation, zinc myristate reacted with the phosphine dangling bonds forming a protective layer on InP core. This treatment led to InP QDs shielded from water and oxygen with a QY of 91 %. [158] Other examples of high quality InP QDs have been prepared by embedding them in mesoporous silica (78 % QY) [159] or by incorporation of metal ions such as neodymium. [160]

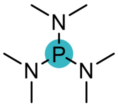
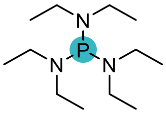
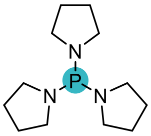
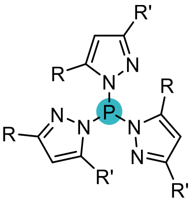
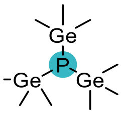
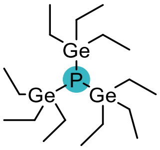
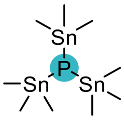
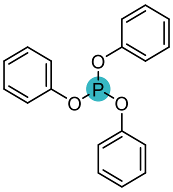
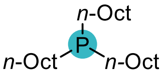
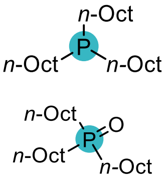
In most of the syntheses of InP, pyrophoric, unstable and toxic tris (trimethylsilyl)phosphine ($P(TMS)_3$, also named PTMS) has been chosen as a phosphorus precursor. Additionally, with an average cost of € 44,000 per mol for $P(TMS)_3$, safer and more economical alternative phosphorus sources are sought. The reactivity of P precursors strongly depends on the energy of their $P-X$ bonds (X = Si, Ge or N, see Table 3). As stated, tris(trialkylsilyl)phosphines are the most commonly used P precursor since Barron et al. first synthesized InP in 1989 from PTMS and $InCl_3$. [161] The P-Si bond dissociation energy (BDE) in PTMS, indeed, is relatively low (around 363 kJ/mol) leading to its high reactivity. [162] On the other hand, such high reactivity is responsible for the fast depletion of the P source causing difficult control on the growth of InP and resulting in wide size distributions. [161,163] Moreover, the use of PTMS restricts the scale-up of InP production due to its sensitivity to temperature homogeneity of the reaction batch. For these reasons, significant efforts are being conducted to develop safer and more sustainable P precursors. One adopted strategy has been the introduction of hindered side chains to PTMS, improving the molecular stability due to the steric effect. Alternatively, many studies are being developed to replace the fragile P–Si bond by stronger P–X bonds (see Table 3). Compounds like P_4, [164] PH_3. [165] and PCl_3. [166] were tested mainly because of their economic potential. However, despite being cheaper than PTMS, these compounds are all highly toxic and are associated with significant hazards. Moreover, the low BDE does not solve the issue of the poor control over nanocrystal size distribution due to fast reaction processes. Considering the thermal stability of the P-Ge and P-Sn bonds compared to the P-Si one, Bawendi and coworkers speculated that replacing PTMS with $P(GeMe_3)_3$ or $P(SnMe_3)_3$ may help in reducing the reactivity and thus facilitate the synthesis of InP. [167,168] The lower reactivity of $P(GeMe_3)_3$ was confirmed but the precursor conversion rate remained too fast to control nanocrystal size effectively.

Another class of phosphorus sources that is gaining more and more interest for the production of InP is the one of aminophosphines. The BDE of the P-N bond, indeed, is much higher than the one of P-Si bond (around 617 kJ/mol) rendering aminophosphines less reactive and able to mediate the growth of InP. [50] The use of these compounds, however, requires the use of reducing agents being the phosphorus in a (+3) oxidation state (versus the (-3) oxidation state in PTMS). The first attempt to synthesize InP QDs using aminophosphines was made by Yang et al., who used tris(dimethylamino)phosphine as the phosphorus precursor and oleylamine as both the coordinating solvent and reducing agent obtaining InP with tunable emission (532–610 nm). [181] Later, Hens et al. adapted this method by reacting aminophosphines with various indium halides and obtaining InP QDs with tunable emission from 550–630 nm, full width at half maximum (FWHM) values of 46–63 nm and QYs of 20–60 %. [174,182] In order to understand the mechanism behind the use of aminophosphines, Dubertret, Mézailles at al. gave some important mechanistic insights underlining the necessity to have an excess of P source due to the reaction reported in Scheme 1. [183] The role of aminophosphines and amines (as reducing agent) was further confirmed in the work of Eychmüller, Weigand at al. [176] InP were synthesized using tri(pyrazolyl)phospanes as the P source and the reaction mechanism was studied. Phosphanes undergo a transamination with oleylamine (OA) to form $HNMe_2$ and $P(OA)_3$ that acted as the effective P source and reducing agent during the formation of the nanocrystals.

The exploration of aminophosphines as PTMS alternative is still an active research area. Being chemically stable, indeed, aminophosphines allow the use of classic Schlenk lines and avoid the need of a glovebox for InP preparation. Moreover, their cost-effectiveness, safety and ease in fine tuning of QDs size and morphology [184] make them one of the most studied alternative P sources nowadays with special attention to tris(dimethylamino)phosphine as can be seen in Table 3. [169–176] Despite this, concerns about their flammability and potential risks for the human health continue to hinder their practical application, especially since their oxidation could lead to carcinogenic phosphoramides. [175] Other phosphorus compounds have been tested, such as triphenyl phosphite, [177] trioctylphosphine, [178] trioctylphoshine oxide [179] or zinc phosphide. [180]

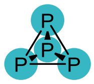
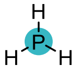
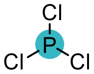
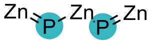
As presented in the previous section, continuous flow technology has a variety of important advantages that can help paving the way towards a more sustainable synthesis of QDs such as rapid mixing, quick heat and mass transfer, accurate control of reagent concentration and temperature, and extreme reaction conditions such as high temperature and pressure. [185,186] In 2009, Nightingale and de Mello reported for the first time the microfluidic synthesis of InP QDs. [187] In their study, a single-capillary and Y-shaped microfluidic devices, were used to synthesize InP QDs. Without optimizing further, the In:Zn ratio, nanocrystals with a QY of 7.0 ± 0.5 % (for the emission at 560 nm) were obtained demonstrating the possibility to produce InP of high quality using continuous flow setups. Later, Bawendi, Jensen et al. developed a microfluidic setup composed of three stages: mixing, aging and sequential injection. [188] The use of high temperatures (up to 300 °C) and high pressures (65 bar) in their system allowed for the use of octane as a supercritical solvent providing excellent mixing and fast diffusivity for producing homogeneous reaction conditions. Also, toluene was used under supercritical conditions for the preparation of InP in a continuous flow reactor. [189] The use of toluene in a flow reaction system provides

Table 3
Examples of alternative P precursors, BDE (bond dissociation energy) of the P-X bond, phosphorus oxidation state and pros/cons of their use.

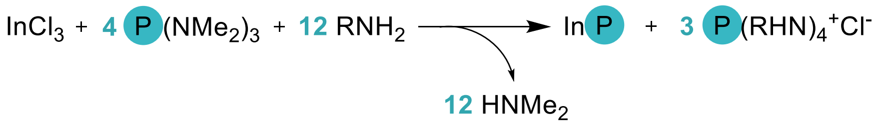
Bond type BDE P oxidation state	Structure	Pros	Cons	Ref.
P-N 617 kJ/mol P(+3)	 Tris(dimethylamino)phosphine	-Cheap, -Low toxicity, -InP of comparable quality as the ones obtained with PTMS.	-Flammable, -Low reactivity: additional reduction agents are necessary, -Potential formation of carcinogenic hexamethylphosphoramide.	[169] [170] [171] [172] [173]
	 Tris(diethylamino)phosphine	-Cheap, -Low toxicity, -InP of comparable quality as the ones obtained with PTMS.	-Flammable, -Low reactivity: additional reduction agents are necessary.	[174]
	 Tris(1-pyrrolidinyl)phosphine	-Cheap, -Simplifies recycling and hazardous waste treatment processes.	-Flammable, -Low reactivity: additional reduction agents are necessary.	[175]
	 Tri(pyrazolyl)phosphanes	-Cheap, -Low-toxic, -Allows to have long-term stable stock solutions.	-Flammable, -Low reactivity: additional reduction agents are necessary.	[176]
P-Ge N.A.[a] P(−3)	 Tris(trimethylgermyl)phosphine	-Stronger P-X bond should allow narrower size distribution, -Larger particles.	-Broad size distributions: precursor conversion rate was still too fast for nanocrystal size to focus, -Presence of byproducts/impurities.	[167]
	 Tris(triethylgermyl)phosphine			
P-Sn N.A.[a] P(−3)	 Tris(trimethylstannyl)phosphine	-Stronger P-X bond should allow narrower size distribution.	-It did not produce InP with sharp absorption features.	[168]
P-O 570 kJ/mol P(+3)	 Triphenyl phosphite	-Non-flammable, -Non-explosive, -Cheap (price is less than one thousandth of PTMS.	-Low reactivity: additional reduction agents are necessary.	[177]
P-C 513 kJ/mol P(−3)	 Trioctylphospine	-Cheap, -Non-toxic.	-Larger particles size than other methods.	[178]
	 Trioctylphospine and trioctylphosphine oxide	-Cheap, -Non-toxic.	-Product has some impurities of free metal.	[179]

(*continued on next page*)

Table 3 (*continued*)

Bond type BDE P oxidation state	Structure	Pros	Cons	Ref.
P-P 460 kJ/mol P(0)	 P_4	-Cheap, -Abundant.	-Spontaneous ignition, -Highly toxic, -Poor control over size distribution due to fast reaction processes.	[164]
P-H 351 kJ/mol P(−3)	 Phosphine (generated in-situ from calcium phosphide)	-Cheap (reduction of the synthesis cost by around three orders of magnitude (vs PTMS).	-Gas (difficult manipulation), -Highly toxic, -Poor control over size distribution due to fast reaction processes.	[165]
P-Cl 356 kJ/mol P(+3)	 Phosphorus trichloride	-Cheap, -Rapid nucleation burst favors the formation of higher-quality NCs.	-Highly toxic, -Poor control over QDs size distribution due to fast reaction processes, -Need of a reductant.	[166]
P-Zn N.A.[a] P(−3)	 Zinc Phosphide	-Cheap, -Stable.	-Need for strong acids, -Involves PH_3 gas formation.	[180]

[a] N.A. = not available



Scheme 1. Synthetic mechanism for the preparation of InP using aminophosphines. The need for an excess of aminophosphines is highlighted.

an opportunity to produce large amounts of QDs simplifying the purification process. The synthesis of shell coated InP/ZnS, was reported from Kim et al. in 2012, using a hybrid flow reactor, based on the combination of a batch-type mixer and a flow-type furnace. [190] The customized setup allowed for the slow addition of ZnS shell precursors, which was critical to suppress the formation of secondary shell nanoparticles through self-nucleation. By optimizing the reaction parameters, InP/ZnS QDs with controllable sizes and colors and with QY up to 37 % were prepared. The synthesis of different core-shell nanocrystals (InP/ZnS, InP/ZnSe, InP/CdS, and InAs/InP) in a multistage microfluidic platform was reported recently from the group of Jensen. [191] In particular, changing the size of the InP core (3.8–4.9 nm), a set of InP/ZnS QDs with tunable emission (554–681 nm) and high QY (32–40 %) were obtained. Also InP/ZnSeS with higher QY (up to 67 %) were obtained with a continuous flow setup in the same year. [192] More recently, a scalable nanomanufacturing strategy for the in-flow synthesis of high-quality InP QDs with multi- stage core growth was developed. A systematic approach to accelerate the synthesis of high-quality heavy metal-free QDs, using a readily reconfigurable network of flow reactor modules was indeed demonstrated in the work by Abolhasani et al. [193] In 2023, Segets and coworkers presented a customized flow reactor for the synthesis of InP QDs using aminophosphines. [194] With the reported approach, InP were obtained with tunable sizes (2.7–3.4 nm) and narrow size distribution. InP/ZnS with QY of almost 31 % were also obtained successfully with their setup.

Despite the advancements and the promising results obtained with InP QDs, the natural scarcity of indium is still a main issue. Indium, indeed, is a rare metal with limited primary sources: lacking a dedicated ore, it is primarily obtained as a by-product of heavy metal smelting, [195] with China as the largest producer and exporter, followed by countries like the Republic of Korea, Japan, Canada, Belgium, and others (see Fig. 9). [196]

To sustain the indium industry, recovering this metal from secondary sources like InP processing waste and end-of-life devices is crucial. While this could supplement limited primary resources, large-scale recycling of waste InP remains still understudied and currently, only limited studies are available in the literature. For instance, Terakado et al. achieved indium recycling from waste InP through a two-step pyrometallurgical process. [197] Initially, waste InP underwent thermal treatment with iron powder, converting it into stable, water-insoluble Fe_3P and metallic indium. Indium was further recovered in the form of $InCl_3$ through chlorination with ammonium chloride, achieving almost full recovery. In another study, Yang et al. introduced a process called “vacuum decomposition-directional condensation” where with a one-step separation (850 °C, 30 Pa, 3.5 h), they retrieved metallic indium and stable red phosphorous with purities of 99.4 % and 98.1 %, respectively. [198] Additionally, a “controlled-pressure pyrolysis–spray condensation” process has been proposed. [199] In this method, InP is decomposed into phosphorus and indium under high-temperature conditions (almost 1050 °C); phosphorus evaporates and is condensed in

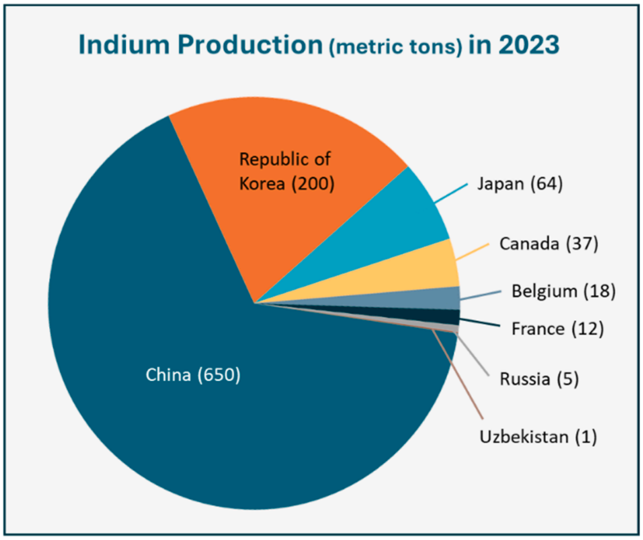


Fig. 9. World refinery production of Indium in 2023 (data from reference [196]).

warm water, while indium remains in the liquid phase. This process achieves zero waste output and successfully recovers indium with 99.59 % purity and a 98 % yield. However, these high-temperature-based process may require costly equipment and significant energy input. Zhao et al. proposed a compact hydrometallurgical method, where InP powder is leached with H_2SO_4 in the presence of H_2O_2, dissolving indium and phosphorus in solution as In^{3+} and PO_4^{3-} ions, respectively. Indium metal was subsequently recovered via cementation using an aluminum plate. [200]

4.3. Carbon quantum dots

CDs are spherical luminescent nanomaterials with diameter smaller than 10 nm and a highly functionalized surface. They have been discovered in 2004 by the purification of single-walled carbon nanotubes. [201] and called “carbon dots” for the first time in 2006 by Sun et al. [202] Nowadays the term CDs comprise different types of carbon-based nanomaterials that have been classified by Valcárcel et al. based on their structure, nature and quantum confinement effect. [203] Principally, below the term CDs we can find:

- Carbon Nanodots (CNDs): quasi-spherical carbon nanoparticles with an amorphous structure and lacking quantum confinement;
- Carbon Quantum Dots (CQDs): spherical carbon nanoparticles with crystalline structure and quantum confinement effect;
- Graphene Quantum Dots (GQDs): nanoparticles composed by π-conjugated sheets presenting quantum confinement effect.

However, the overall structure and performance of each class of CDs are very similar and they will be discussed together in this section. CDs exhibit several unique properties, including high water solubility, nontoxicity, biodegradability, tunable fluorescence, ease of modification, and chemical stability. These characteristics enable CDs to be applied across various fields, such as biomedical (e.g., bioimaging [204] and drug delivery [205]), sensor development, [206] optoelectronic devices, [207] and photocatalysis. [208,209] Furthermore, CDs can be synthesized using low-cost techniques from a broad range of inexpensive and abundant precursors, including biowaste, making them very appealing to replace other more expensive materials. Lately a large number of different techniques have been developed in order to synthesize CDs starting from a wide array of different carbon precursors. These synthetic pathways can be divided in two main families, namely the top down approaches and the bottom up ones. Top down methods involve the breakage of large carbogenic structures such as graphite, graphite layers, or carbon nanotubes, while in the bottom up carbon NPs are obtained by carbonization of small organic molecules.

CDs are primarily composed of carbon, hydrogen, and oxygen, with their specific ratios dependent on both the precursor used and the chosen synthesis method. To achieve CDs with different properties, additional elements can be introduced during synthesis. Nitrogen is the most commonly used doping element, as it enhances the QY. [210,211] Other dopants, such as sulfur, can widen the band gap, [212] while phosphorus can create green-emitting dots, which are important for bioimaging applications. [213] Also the morphology of the carbogenic core in CDs can vary depending on the synthetic method and reaction conditions. For instance, CDs produced via top down approaches typically have a highly crystalline core with well-defined layers, whereas CDs synthesized through bottom up methods often exhibit more ambiguous morphologies. Recent studies have aimed to better understand the structure of CDs and how the synthetic process influences their characteristics. [214] Overall, top down approaches generally result in GQDs, while bottom up synthesis methods are primarily used for producing CQDs and CNDs. [215–217]

CDs are generally considered as a non-toxic alternative to classic QDs. However, the great variety in structures and composition of this class of nanomaterials lead to different results also in terms of toxicity. Perosa et al., for example, prepared CDs starting from glucose, ascorbic acid and fructose to be used in a drug delivery system and tested their toxicity towards HeLa cells at different concentrations. From this study it resulted clear that the CDs derived from glucose resulted non-toxic, the one from ascorbic acid were biocompatible only at concentration <250 μg/mL, and the one obtained from fructose were highly toxic. [218] Kuznietsova et al. tested CDs with different surface groups for *in vitro* and *in vivo* toxicity. For all the type of tested CDs no *in vitro* toxicity was observed. Depending on the functional groups, however, some *in vivo* toxicity was highlighted with immediate or delayed responses. [219] If toxicity studies on CDs are still ongoing, the sustainability of new synthetic methodologies renders them a real alternative to classic QDs.

The production methods, indeed, are nowadays developing with a special focus on green and safer alternatives, particularly on the use of waste materials as the carbon source. The use of waste can be applied mainly in bottom up approaches but some example of top down synthesis using candle soot, [220] vehicle exhaust soot [221] or coal tar pitch. [222] were also reported. Although the use of recyclable and reusable waste materials aligns to some extent with the concept of green chemistry, the use of strong acids or oxidants in top down approaches remains a significant obstacle to further commercialization. In contrast, bottom up methods are generally simpler and safer. Among these, the hydrothermal method is the most commonly employed for synthesizing CDs from green sources due to its simplicity, shorter reaction time, mild reaction conditions, and suitability for large-scale synthesis. [223] The hydrothermal method is considered environmentally friendly because it uses non-toxic materials and operates under non-polluting synthesis conditions. Table 4 provides examples of CDs synthesized from various waste types (e.g., vegetal, animal, plastics, and others).

Vegetal waste matrices are among the most used as carbon precursors. For example, Shang, Che et al. obtained CDs from agricultural waste corn cob by an ionic liquid mediated hydrothermal treatment. [225] The as obtained CDs resulted to have uniform size distribution with a diameter of 1.5 ± 0.5 nm, a clear crystal lattice and a QY of 15.5 % with emissions at 405 and 425 nm. The corn cob-based CDs were then used to prepare a fluorescent probe for melamine detection together with silver NPs. A plethora of different plant-based waste have been used for the production of carbon NMs, such as different types of leaves, [224,228,233,235] orange or lemon peels, [226,232] peanut shells, [229] pea sesame, [227] konjac flour, [230] garlic [231] or banana pseudo-stems. [234] Biomass is a complex matrix that can contain heteroatoms such as N and S, that can lead to a self-doping effect to CDs. Therefore, the characteristics of waste-derived CDs heavily rely on the choice of the carbon precursors. For instance, naturally nitrogen doped CDs were obtained *via* hydrothermal treatment of bass scale showing a significant amount of nitrogen (C:N = 3.21). [237] These CDs were used as a photocatalyst for the reduction of methyl viologen showing high photoelectron transfer ability. The same bass scale-derived CDs were applied successfully as photocatalyst also for the continuous flow degradation of pollutants and for the preparation of UV shields. [245,246] Among other animal-based waste that have been employed for the preparation of CDs we can find prawn and crab shells, [236,238] milk [239] and leather scrap. [240] Lately, the upcycling of plastics into CDs is gaining interest in the scientific community. An interesting case was reported by Ramadi at el. whom used a single-step hydrothermal process to convert polyethylene-based plastic bags and polypropylene-based surgical masks into CDs with a 96 % production yield. [243] Polystyrene and polylactide were also effectively employed as carbon source. [241,244]

As already mentioned for InP, CF chemistry is a valuable tool to achieve more sustainable production of NMs (see Table 5). Although the potential of CF synthesis for CDs is significant, the first report on this topic only appeared in 2014, [247] and to date, only a limited number of studies have been published. The use of microreactors provides high control over the average size of CDs by regulating the flow rate and

Table 4
List of waste used as C source for CDs preparation, the applied synthetic method, size and QY of the obtained CDs and their application field.

Type of waste	Carbon Source	Synthetic method	Size (nm)	QY (%)	Application	Ref.
Vegetal	Catharanthus roseus leaves	Hydrothermal	5	28.2	Sensing and Bioimaging	[224]
	Corncob	Hydrothermal	1.5	1.8	Melamine detection	[225]
	Orange peel	Hydrothermal	2.03	N.A.[a]	Electrode	[226]
	Pea sesame	Hydrothermal	3.1–4.3; 1.7–2.8	2.5	Labelling	[227]
	Tobacco leaves	Hydrothermal	6.3	13.7	Fluorescent probe	[228]
	Peanut shell	Pyrolysis	1.8–4.2	10.6	Probe for copper ion	[229]
	Konjac flour	Pyrolysis	3.4	22	Cell imaging	[230]
	Garlic	Microwave	20	54	Antioxidant	[231]
	Lemon peel	Hydrothermal	1–3	14	Detection and Bioimaging	[232]
	Wild lemon leaves	Microwave assisted pyrolysis	4.08	7.2	Tetracycline detection	[233]
	Banana plant pseudo-stems	Hydrothermal	1–3	48	Bioimaging	[234]
	Date palm fronds	Hydrothermal	< 10	33.7	Photocatalysis	[235]
Animal	Prawn shells	Hydrothermal	4	9.0	Detection of copper ions	[236]
	Bass scale	Hydrothermal	10	6.0	Photocatalysis	[237]
	Blue crab shell	Hydrothermal	6	14.5	Detection of ceftriaxone	[238]
	Milk	Hydrothermal	2	8.6	Wearable optoelectronics and security labelling	[239]
	Leather Scrap	Hydrothermal/ solvothermal	2.5–10.0	1.05 – 5.14	Anti-counterfeiting	[240]
Plastic	Polylactide	Hydrothermal	3	N.A.[a]	N.A.[a]	[241]
	PET	Hydrothermal	12.9	9.1	Metal ion sensing	[242]
	Polyethylene and polypropylene	Hydrothermal	1–8	14.6–16	Anticounterfeiting agents	[243]
	Expanded polystyrene	Solvothermal	2.8–5.2	20	Detection of Au ions	[244]
Others	Candle soot	Combustion	< 2	< 1	Possible fluorescence labels	[220]
	Vehicle exhausted soot	Acid reflux	4	3.0	Tartrazine sensing	[221]
	Coal tar pitch	Hydrothermal	1.9 – 5.8	N.A.[a]	Photocatalysis	[222]

[a] N.A. = not available

Table 5
Examples of continuous flow preparation of CDs in different systems. The nature of the precursors, solvent, type of reactor and size and QY of the CDs are reported. HP = High Pressure; SS = Stainless Steel.

Precursors	**Solvents**	**Reactor type**	**Size (nm)**	**QY (%)**	**Ref.**
Citric acid, L-cysteine	Water	Coil micro-reactor	2.8	68.2	[253]
Citric acid, N sources	Water	Co-fired ceramic micro-reactor	2.2–4.3	< 0.77	[250]
Glucose, *p*-sulfonic acid, calix4arene	scH$_2$O	HP scH$_2$O reactor	1.7	0.25	[254]
Glucose	scH$_2$O	HP scH$_2$O reactor	2.3	0.3	[255]
Glucose, ammonia	scH$_2$O	HP scH$_2$O reactor	4.6	9.6	[256]
Citric acid, ethylene-diamine	Tetra ethylene alcohol, water	SS coiled micro-reactor	10	26.95	[257]
Ethanolamine, phosphoric acid	Water	SS capillary micro-reactor	2.6	N.A.	[258]
Milk/soy milk, orange/ watermelon juice	Water	SS capillary micro-reactor	2.17	12.53	[259]
Ascorbic acid, diethylene-triamine	Water, ethylene glycol	Poly (dimethyl-siloxane) microfluidic chips	4.0	19.2	[260]

[a] N.A. = not available

adjusting the reactor's length, size, and shape. [248] For instance, shorter residence times are associated with smaller particles, [249] while reduced reactor diameters result in a narrower size distribution of CDs due to more uniform and faster heat transfer. [250,251] Furthermore, microreactors are energy-efficient, minimize waste production, and allow for safer handling of extreme temperature and pressure conditions. [252] Their use has also been shown to enhance the QY of CDs. [131]

Table 5 summarizes recent studies on CF synthesis of CDs with the used precursors, solvents, reactor type and characteristics of the obtained dots. A significant example has been given by Chen, Xu and coworkers who used a coiled microreactor to continuously produce CDs from citric acid and L-cysteine in water with a residence time of 10 min. [253] The obtained NPs resulted to be nitrogen and sulfur-doped and demonstrated a bright luminescence with a high QY of 68.2 %. These CDs demonstrated high sensitivity for cadmium ion (Cd^{2+}) detection, achieving a detection limit of 0.079 ppb and excellent selectivity. As evidenced in Table 5, water is the preferred solvent also for the continuous flow production of carbon NMs resulting in the so called Continuous Hydrothermal Flow Synthesis (CHFS). Also, water in supercritical conditions (scH$_2$O) have been used successfully in a number of studies thanks to the flexibility in controlling its density and dielectric constant. For instance, Kellici and coworkers utilized scH$_2$O at 450 °C and 248 bar to produce sulfur-doped CDs from glucose and *p*-sulfonic acid calix4arene [254] and non-doped CDs from bare glucose [255] resulting in small CDs (1.7 and 2.3 nm respectively) with low QY (0.25 and 0.3 %). Glucose has been employed as a carbon precursor together with ammonia in scH$_2$O, also from another group obtaining N-doped CDs with higher QY (9.6 %). [256]

All the examples using scH$_2$O as solvent used a similar setup as schematized in Fig. 10. The setup employed three feedstock streams: one for water, one for the carbon source and one for the eventual doping agent. Water was heated at 450 °C using a heater and its pressure was kept at 24.8 MPa with a back pressure regulator (BPR). A "T" junction was used to mix the two precursors and the solution met the scH$_2$O stream in a counter current mixer. After a cooling step the CDs were collected.

Less harsh conditions were also successfully employed to obtain CDs using stainless steel microreactors. [257–259] Lin et al. obtained nitrogen and phosphorus-doped CDs in CF applying temperatures from 110 to 170 °C and at ambient pressure. [258] In an interesting study from 2023, CDs were synthesized with a microfluidic method using as precursor food waste (milk or fruit juice). [259] Also at relatively low temperature (120 °C) the obtained dots resulted to have acceptable QY (12.5 %) and good stability demonstrating the possibility to adopt green

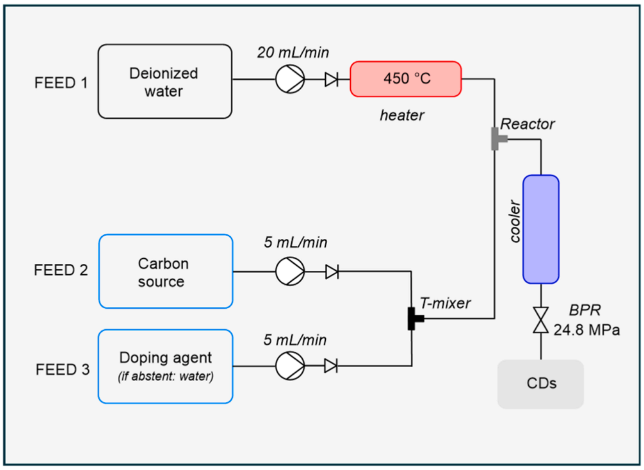


Fig. 10. Continuous flow setup for the synthesis of CDs in scH_2O.

approach for the synthesis of carbon nanomaterials. Even lower temperatures were demonstrated to be efficient for CF CDs production in the work from Jiang et al. [260] Three-dimensional microfluidic chips in poly(dimethylsiloxane) were fabricated using soft lithography and used for the obtainment of CDs from ascorbic acid and diethylenetriamine at 90 °C. The resulting CDs had uniform particle size and QY of 19.2 % and were used for the detection of cefquinome residues.

4.4. Other alternatives to II-IV QDs

Besides InP QDs and CDs, numerous less toxic alternatives to classic II-IV QDs are under development. This section briefly discusses some examples, providing an overview of possibilities to enhance the safety of QDs.

One highly promising class includes I-III-VI QDs, composed of group I (Cu, Ag), group III (Al, Ga, In, Tl), and group VI (S, Se, Te) elements, such as $AgInS_2$, $CuInS_2$, and $CuInSe_2$. I-III-VI QDs exhibit advantageous features like tunable emissions across the visible to near-infrared (NIR) spectrum, high absorption coefficients, large Stokes shifts, long photoluminescence lifetimes, and low toxicity. [261] For instance, $CuInS_2$, with a narrow bandgap energy of 1.45 eV, emits in the NIR region and its emission peak can be easily tuned by adjusting the reaction temperature. [262] The interesting properties of I-III-VI QDs make them suitable for applications in light-emitting devices, [263] solution-processable solar cells, [264] bioimaging, [265] and photocatalysis. [266] The low toxicity of these materials has been confirmed in several studies. Pérez-Donoso et al., for example, synthesized $CuInS_2$ via a biomimetic method using $CuSO_4$ and $InCl_3$ as precursor salts, with glutathione as sulfur donor and stabilizer. [267] The synthesized $CuInS_2$ showed no cytotoxicity in *E. coli* even at concentrations of 1000 mg/mL. Similarly, no effect on epithelial cell viability was observed at concentrations ranging from 50 to 200 mg/mL.

However, synthesizing I-III-VI semiconductors remains challenging due to the need to balance the reactivity of multiple precursors for ternary nanocrystal nucleation and growth and for the presence of competing pathways that can lead to the formation of binary systems. While the hot injection method has yielded the best results, other approaches such as hydrothermal and microwave-assisted synthesis, thermal decomposition, and photochemical or ultrasound methods have also been explored to produce comparable quality I-III-VI QDs. [261]

Another promising class of sustainable and safe nanocrystals is the one of silicon-based QDs. Bulk silicon, an indirect band-gap semiconductor with an energy gap of 1.12 eV, exhibits negligible photoluminescence due to slow electron-hole radiative recombination. [21] However, nanometer-sized silicon dots display efficient and tunable optical properties due to quantum confinement effects. [268] The size, shape, and surface passivation of the nanoparticles significantly influence these optical characteristics. Thanks to these features, silicon QDs can be suitable for applications such as light-emitting diodes [269] and solar cells. [270] Another interesting application of Si-QDs is their use for quantum computing. Electron spins in Si-QDs, indeed, attracted a lot of interest as a platform of quantum computation with high-fidelity universal quantum control, long coherence time, capability of high-temperature operation, and potential scalability. [271]

In order to assess the toxicity of Si-QDs, a comparative study of Si-QDs, CdSe, and CdSe/ZnS QDs using Gram-negative and Gram-positive bacteria has been performed. [272] The results showed that Si-QDs are benign and do not associate with cell membranes, whereas CdSe QDs exhibit significant toxicity. Core-shell CdSe/ZnS QDs were found to be less toxic than CdSe cores but still more toxic than Si-QDs, highlighting the positive impact of a protective shell. Moreover, it has been demonstrated that the rapid degradation of Si-QDs in biologic media leads to the formation of non-toxic salicylic acid, which is advantageous for biological imaging. [261]

Despite these advantages, synthesizing high-quality Si-QDs remains challenging, and several researchers nowadays focus on the development of new synthetic pathways and surface passivation methods. [273] Si-QDs have been prepared using various methods, including chemical reduction and physical techniques. Chemical reduction methods employ reducing agents like $LiAlH_4$ [274] and sodium naphthalene. [275] Physical approaches include the thermal processing of hydrogen silsesquioxane, [276] or ion implantation. [277] These methods, however, face several challenges such as high synthesis costs, which limit their scalability and industrial applicability. Moreover, using chemical reduction methods, the optical properties of the resulting colloidal Si-QDs are often constrained leading only to blue or green luminescence, due to defect states on the surface of the dots. These surface defects significantly impact the quality and utility of the synthesized nanocrystals, underscoring the need for improved synthesis and passivation techniques. More sustainable, green and cost-effective synthetic approaches have also been recently developed. For example, Hernandez-Abril and coworkers developed a preparation method for Si-QDs using leaf extract of *Ocimum basilicum var. purpurascens* as an organic silicon-reducing agent and 3-aminopropyltriethoxysilane as a precursor. [278] Rice husk has also been used as an efficient bio-source of silica for the preparation of Si-QDs minimizing the environmental impact of classical syntheses. [279–281]

In addition to the discussed classes of less-toxic QDs, a plethora of different families are being developed highlighting the interest of the scientific community in replacing II-IV QDs. Some remarkable examples are represented by lead-free halide perovskite, [282,283] zinc chalcogenides [284,285] or boron nitride QDs. [286,287]

5. Summary and outlook

For decades, semiconductor quantum dots have emerged as a pivotal technology in materials science. Their versatile structure and tunable band gap enable their use in diverse applications ranging from bioimaging and optoelectronics to photocatalysis. Despite extensive research, knowledge gaps persist regarding the regulations and potential risks of QDs for human health and the environment. Therefore, this review first aims to clarify the European Union's regulatory framework on NMs, with a particular focus on QDs. Additionally, it discusses the factors influencing QDs toxicity and concludes by exploring safer and more sustainable methods and alternatives for their preparation.

The regulatory landscape surrounding NMs has evolved alongside advancements in nanotechnology and nanoscience. The progress made so far is a result of collaborative efforts among governments, international organizations, researchers, and academics, all of whom are genuinely concerned about the possible harmful implications of these materials. However, the diversity of NMs has made the regulatory framework development a complex and ongoing process. Despite current achievements, there remains a substantial path ahead. Beyond merely establishing methodologies and techniques, NMs regulations must aim to ensure the safety and usefulness of nanotechnological products. In doing so, they support public trust, scientific progress, improved quality of life, and environmental protection. A significant challenge lies in the lack of specific legislation targeting QDs. While considered a subset of NMs, QDs represent a distinctive class with varied compositions and properties. Given the rapid growth in QDs research and applications, developing specialized regulations is imperative. In the broader scope of science and technology, advancements continually contribute to human progress, and nanoscience is no exception. Consequently, the trend toward incorporating NMs, including QDs, will likely persist as it has over the past two decades. The necessity of establishing comprehensive legislation to safeguard consumer and worker safety will only become more apparent over time. In parallel, educational programs are essential to communicate the science behind NMs and QDs, raising awareness about their uses, management, and disposal across all sectors of society.

Concerning classic QDs, such as Cd-based ones, achieving universal approval is very challenging due primarily to stringent restrictions in certain jurisdictions (e.g. RoHS in the EU). However, some regulations allow exceptions for materials with significant performance advantages, provided that adequate safeguards are implemented. In particular, several regulations limit the amount of Cd in products but do not ban its use (e.g., regulation on batteries or RoHS regulation). This suggests that Cd-based QDs might still be viable for specialized applications where their unique properties are critical, and where robust mitigation strategies are in place. A critical challenge in this context is achieving a balance between the performance benefits of classic QDs and their environmental and health trade-offs. While Cd-based QDs are highly valued for their superior optical properties, their toxicity necessitates advanced strategies to mitigate risks.

Undesirable toxicity effects have, indeed, significantly hindered the practical application of QDs. Moreover, the structural and compositional variability of QDs complicates toxicity studies, underscoring the need for standardized protocols. Reducing QDs toxicity should be a primary goal for future research. In particular, concerning classic Cd-based QDs, different strategies such as core-shell coating and precise control over size, concentration, and charge should be further studied and adopted. While these approaches can significantly reduce the release of toxic Cd cations and improve biocompatibility, such measures may not completely eliminate the inherent risks associated with Cd-based materials.

Developing safer alternatives to Cd-based QDs become then even more crucial. Indium phosphide (InP) and carbon dots (CDs) offer promising heavy metal-free and metal-free substitutes, respectively, especially when synthesized using sustainable materials and continuous flow methods. However, for InP and CDs to replace Cd or Pb-based QDs fully, they must improve in terms of luminescence and quantum yield. Advancements in synthetic technologies for producing high-quality, non-toxic QDs at scale are also essential, along with more rigorous toxicity testing for InP and CDs. A summary of the main conclusions and future perspectives for the sustainable production and use of QDs is represented in Fig. 11.

Irrespective of the type of QDs, the effective recycling of these materials remains a key concern in today's global context. Heavy metals or rare metals make recycling efforts challenging because of health risks and environmental regulations associated with their disposal and recovery. Recycling Cd-based QDs involves complex chemical separation processes due to Cd toxicity and requires stringent disposal or containment measures to avoid environmental contamination. This makes recycling Cd-based QDs costly and heavily restricted by Cd usage regulations. In contrast, InP and CDs present more sustainable, low-toxicity alternatives, although they involve distinct challenges related to performance and scalability. For InP, recycling primarily focuses on material recovery, driven by the scarcity of indium. Moreover, current recycling methods are expensive, and manufacturing new QDs is often more cost-effective than recycling. Developing cost-efficient and scalable recycling methods is therefore a key goal to ensure the sustainable production and use of semiconductor nanocrystals. Potential alternatives to traditional acid leaching or high-temperature and high-pressure methods include biodegradation using microorganisms and photodegradation but the research on these areas is still in its infancy. Moreover, as regulations on electronic waste and hazardous materials increase, there may be more incentives or requirements for manufacturers to invest in QDs recycling. A comparison of Cd-based QDs, InP and CDs regarding their environmental impact and toxicity, their efficiency and ease in recycling is depicted in Fig. 12.

Depending on the application scenarios, InP and CDs can each bring unique advantages and challenges. InP QDs are highly valued for their tunable optical properties, which make them ideal for display technologies and bioimaging applications that require specific wavelengths. They also exhibit good photostability, maintaining consistent performance under prolonged light exposure. However, as already mentioned, the synthesis of high-quality InP QDs is complex and the presence of

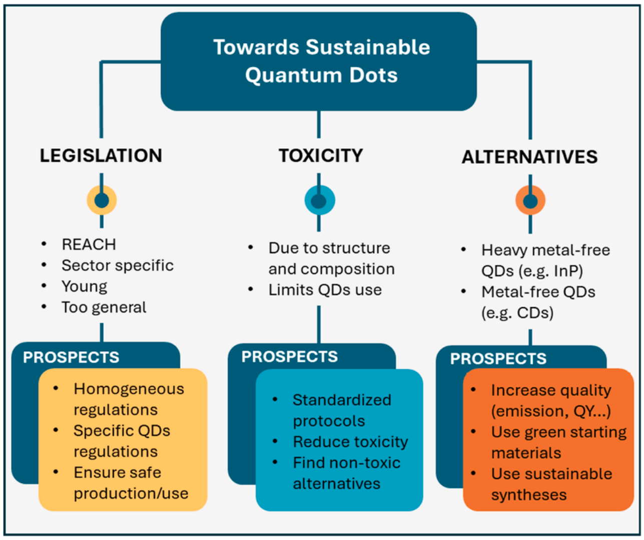


Fig. 11. Conclusions and main prospects for sustainable use and preparation of QDs.

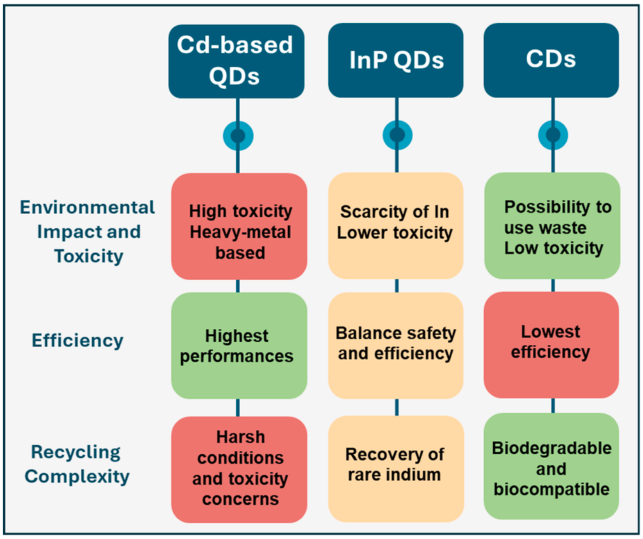


Fig. 12. Comparison of traditional Cd-based QDs, InP and CDs for their environmental impact and toxicity, efficiency and recycling complexity.

surface defects can reduce their efficiency in optoelectronic applications. On the other hand, CDs are particularly suitable for biomedical applications due to their excellent biocompatibility and low toxicity. They are also highly versatile, as their surfaces can be easily functionalized to suit various needs. CDs benefit from cost-effective and straightforward synthesis processes, often utilizing abundant carbon sources. Nonetheless, they tend to exhibit lower QY compared to InP QDs, which can limit their use in applications requiring high emission efficiency. Furthermore, CDs typically absorb in the ultraviolet to blue spectrum, restricting their use in scenarios that demand absorption at longer wavelengths.

In conclusion, the advancement and tremendous potential of nanotechnologies, particularly QDs, necessitates the development of an extensive regulatory framework to govern their production and use, with an emphasis on mitigating potential toxicity. Strategies to control and minimize adverse effects are a key area of research and will likely intensify interest in the field of QDs. However, the successful integration of QDs into society depends on a collaborative approach involving not only scientists and researchers but also policymakers, regulatory bodies, and industry stakeholders. Only through coordinated efforts across these sectors it is possible to ensure that the development of QDs proceeds safely and sustainably, thereby maximizing their benefits for human progress and environmental health. Fostering open dialogue and continued investment in education will be essential to raise awareness and build a foundation for responsible innovation in this rapidly evolving field.

CRediT authorship contribution statement

Campalani Carlotta: Writing – original draft, Conceptualization. **Monbaliu Jean-Christophe M.:** Writing – review & editing, Supervision, Conceptualization.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jean-Christophe M. Monbaliu reports financial support was provided by WEL Research Institute (WEL-T-CR-2023 A – 05). If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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