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REVIEW

Cyclodextrins, from molecules to applications

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Abstract
Cyclodextrins are among the most remarkable macrocyclic molecules with significant theoretical and practical impacts in chemistry and biology. Cyclodextrins belong to the family of cage molecules due to their structure, which is composed of a hydrophobic cavity that can encapsulate other molecules. Indeed, the most characteristic feature of these compounds is their ability to form inclusion complexes with various molecules through host–guest interactions. This is at the origin of many applications. It is well known and widely reported in the literature that cyclodextrins and their derivatives have a wide variety of practical applications including pharmacy, medicine, foods, cosmetics, toiletries, catalysis, chromatography, biotechnology, nanotechnology, and textile industry. Cyclodextrins are also the object of numerous fundamental studies. In this review, we chose to highlight selected works on cyclodextrins published over the last 5 years by different research groups. The main objective is to summarize some of the recent developments related to the applications of cyclodextrins.

Keywords Cyclodextrin · Applications · Recent overview

Cyclodextrins: an introduction

This article is an abridged version of the chapter published by Crini et al. (2018) in the series Environmental Chemistry for a Sustainable World.

Cyclodextrins are cyclic oligomers obtained from the enzymatic degradation of starch, one of the most essential polysaccharide in the nature. Cyclodextrins make up a family of cyclic oligosaccharides. They are composed of six or more D-glucopyranoside units linked in α(1–4), like in amylose, a component of starch. Typical native cyclodextrins contain six, seven, or eight glucose units and are denoted α-, β-, and γ-cyclodextrins, respectively (Fig. 1). They are produced in a highly pure form at an industrial scale. Cyclodextrins are hollow, truncated-cone-shaped molecules made up of several glucose units linked together covalently by oxygen atoms and held in shape via hydrogen bonding between the secondary hydroxyl groups on adjacent units at the wider rim of the cavity. Indeed, cyclodextrins are ring molecules and they are toroidal or cone-shaped (Fig. 1).

This review is dedicated to the memory of Professor Benito Casu (*Istituto di Chimica e Biochimica G. Ronzoni*, Milan, Italy).

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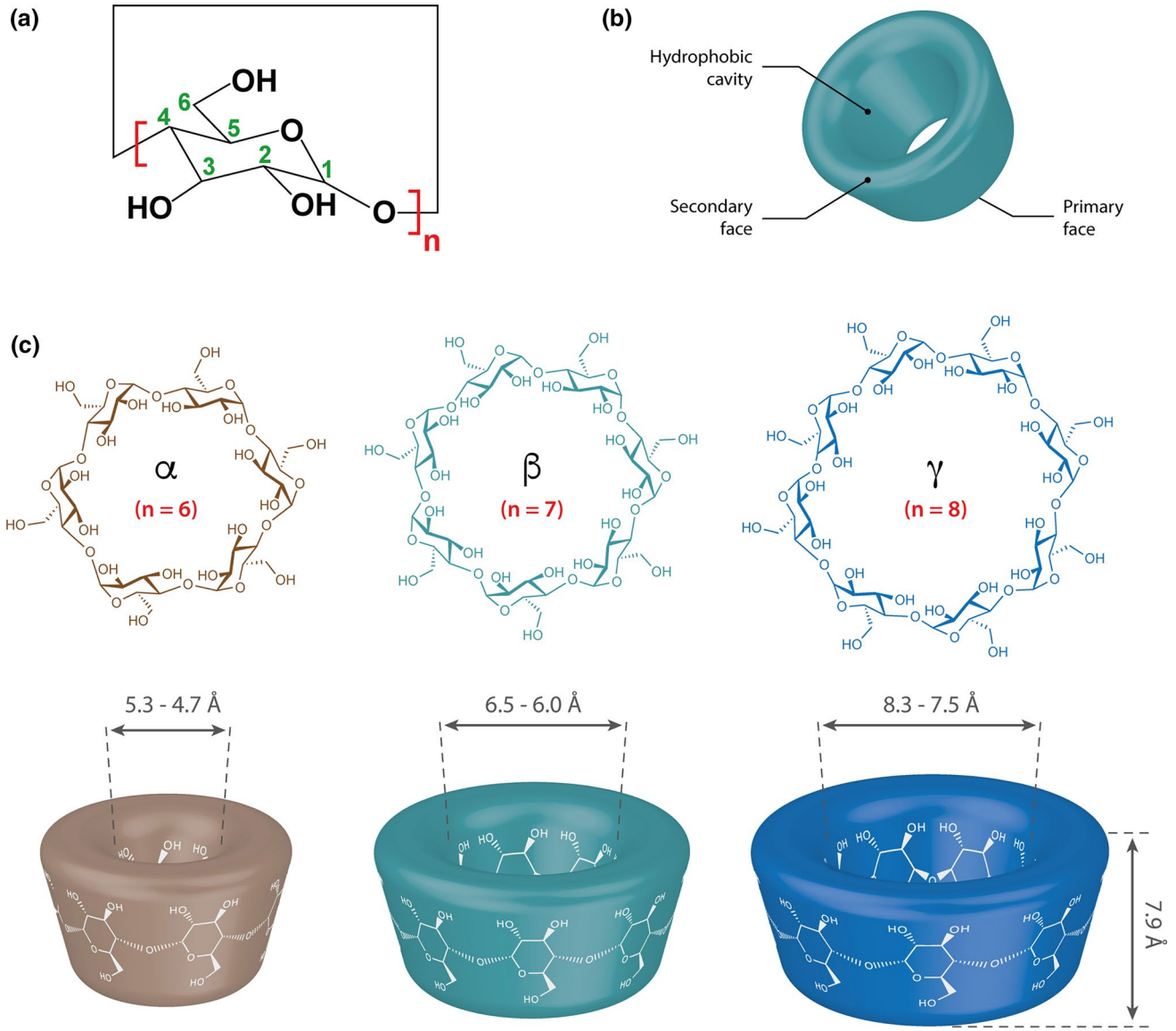

Fig. 1 Schematic representations of **a** the general chemical structure and **b** the tridimensional structure of cyclodextrins, and **c** chemical structure and dimensions for α-, β- and γ-cyclodextrin ($n = 6$, 7 and 8, respectively)

β-Cyclodextrin (β-CD) is the most studied and most frequently used, based on its cheapness, availability, and complex-forming capacities toward a large range of substances. Indeed, as others cyclodextrins, the most characteristic feature of the β-CD molecule is its ability to form inclusion compounds with various substances through host–guest

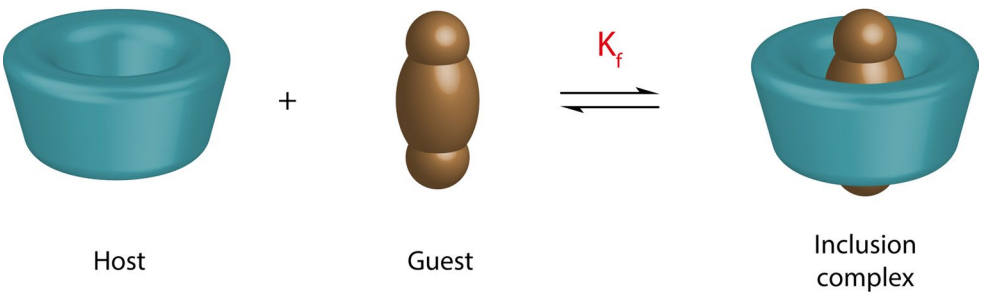

Fig. 2 Schematic representation of the inclusion phenomena between a cyclodextrin molecule (the host) and a solute (the guest) to form solute–cyclodextrin complexes

interactions (Fig. 2). Its central cavity which is composed of seven glucose units is hydrophobic when the external part is hydrophilic because the presence of 21 hydroxyl groups. The core of its structure can trap or encapsulate other substances. These remarkable encapsulation properties can modify and/or improve the physical, chemical, and/or biological characteristics of the guest molecule (Szejtli 1988; Morin-Crini et al. 2015).

Although they have been known for 127 years, cyclodextrins only really took off in the 1980s with the first applications in the chromatography, pharmaceutical and food industries (Crini 2014). Actually, applications are found in practically all sectors of industry (Morin-Crini et al. 2015). The literature on cyclodextrins is vast and spread across different disciplines: Chemistry, Biochemistry, Health Science, Agriculture, etc. Interesting books and book chapters on the different aspects of cyclodextrins such as the preparation, description, characterization, properties, chemistry, and derivatives for these disciplines can be consulted (Table 1). Figure 3 demonstrates how the number of publications increased using cumulative numbers of 5-year periods. In 2015, with an average of 4.5 papers and 2.5 patent applications daily, the literature data show that cyclodextrin research and development is still in the focus (Source: Cyclodextrin News, Cyclolab).

The main objective of this review is to summarize some of the recent developments related to the applications of cyclodextrins. To do this, we chose to highlight selected works on cyclodextrins published over the last 5 years by different research groups in order to have an update overview. Of course, this review is not exhaustive. Readers interested in cyclodextrin topics should refer to the library database "Cyclodextrin News" from Cyclolab, Ltd. (Hungary), which is a periodical collecting all of the cyclodextrin papers, proceedings, patents, conferences and lectures.

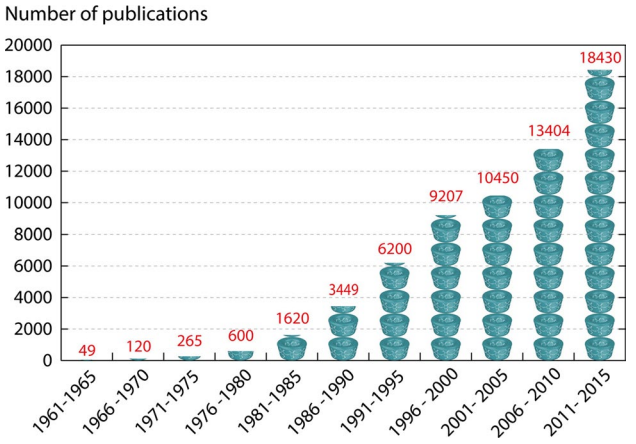

Fig. 3 Number of cyclodextrin-related publications including papers, patents, and conferences (source: Cyclodextrin News, Cyclolab)

Table 1 Selected examples of books on cyclodextrins

Year	Title	Reference
1978	Cyclodextrin chemistry	Bender and Komiyama (1978)
1982	Cyclodextrin and their inclusion complexes	Szejtli (1982)
1984	Inclusion compounds	Atwood et al. (1984)
1987	Cyclodextrins and their industrial uses	Duchêne (1987)
1988	Cyclodextrin technology	Szejtli (1988)
1991	New trends in cyclodextrins and derivatives	Duchêne (1991)
1994	Cyclodextrin in pharmacy	Frömming and Szejtli (1994)
1996	Comprehensive supramolecular chemistry. Volume 3: Cyclodextrins	Szejtli and Osa (1996)
2000	Principles and methods in supramolecular chemistry	Schneider and Yatsimirsky (2000)
2002	Introduction to supramolecular chemistry	Dodziuk (2002)
2004	Encyclopedia of Supramolecular Chemistry	Atwood and Steed (2004)
2006	Cyclodextrins and their complexes—Chemistry, analytical methods and applications	Dodziuk (2006)
2010	Use of cyclodextrin polymers in separation of organic species	Kozlowski and Sliwa (2010)
2011	Cyclodextrins in Pharmaceutics, Cosmetics and Biomedicine. Current and Future Industrial Applications	Bilensoy (2011)
2012	Applications of Supramolecular Chemistry	Schneider (2012)
2015	*Cyclodextrines*: *Histoire, propriétés, chimie et applications* (in French)	Morin-Crini et al. (2015)
2016	Encapsulations	Grumezescu (2016)
2018	Cyclodextrin: Fundamentals, Reactivity and Analysis	Fourmentin et al. (2018)

Cyclodextrins: an update 2012–2016—present situation, trends and outlook

Self-association of cyclodextrins

It is now well known that cyclodextrin molecules can adopt various types of assembly modes in aqueous solution as well as crystal structures (Gonzalez-Gaitano et al. 2003). New micro- and nanostructures including aggregates, formed by the self-assembly of cyclodextrins, have been useful, particularly in the fields of supramolecular chemistry, materials science, pharmacy including formulation and drug delivery, and medicine as reported by Kurkov and Loftsson (2013), Simoes et al. (2014), Ryzhakov et al. (2016), and Oliveri and Vecchio (2016).

The most important property of the cyclodextrins is the ability to establish specific interactions (molecular encapsulation) with various types of molecules through the formation of non-covalently bonded entities, either in the solid phase or in aqueous solution. However, cyclodextrins are not only able to form these host/guest inclusion complexes but also non-inclusion complexes (Gonzalez-Gaitano et al. 2003; Loftsson et al. 2004; Messner et al. 2010; Moya-Ortega et al. 2012; Kurkov and Loftsson 2013). The hydroxyl groups present on the outer surface can form hydrogen bonds with other molecules which make them able, just like dextrins, non-cyclic oligosaccharides, and polysaccharides, to form complexes (molecular structures) with lipophilic substances insoluble in water. In pure aqueous cyclodextrin solutions, CD molecules self-assemble to form nanoparticles with diameter from about 20 to 200 nm (Loftsson et al. 2004; He et al. 2008). At low concentrations (below 1% w/v), the fraction of CD molecules forming such aggregates is insignificant but the aggregation increases with increasing cyclodextrin concentration. Another possibility that is mentioned is the formation of aggregates able to dissolve water-insoluble lipophilic molecules (structures similar to micelles). The CD/drug complex aggregates are frequently from 100 to 4000 nm in diameter or in the nanoparticle and small microparticle size range. The complexes are kept together by weak hydrogen bonds and hydrophobic forces and dissociate readily upon media dilution. Such systems are called cyclodextrin nanostructures, nanoassemblies, or self-aggregates.

It has been observed, in pharmaceutical applications, that other types of cyclodextrin complexes such as non-inclusion complexes are also participating in CD solubilization of poorly soluble drugs (Bilensoy 2011; Kurkov and Loftsson 2013; Morin-Crini et al. 2015). There are some indications that formation of CD/drug complex aggregates might play an important role in CD enhancement of drug bioavailability. The cyclodextrin aggregates present the ability to form complexes, and nanosized aggregates and nanotube-type host/guest architectures can be envisaged. This is a generally unexplored domain and often causes controversies since the results obtained are closely dependent on the technique used, as pointed out by Ryzhakov et al. (2016). Another problem discussed in the literature is the stability of the cyclodextrin aggregates, in particular when these systems are used as drug delivery systems. The publications of Loftsson's group summarized the most important features and the main conclusions of these cyclodextrin-based aggregates (Fülöp et al. 2012; Kurkov and Loftsson 2013; Ryzhakov et al. 2016; Saokham and Loftsson 2017). The use of CD-based nanoaggregates in oral drug delivery could be a promising strategy to improve the bioavailability of poorly soluble drugs. Much work is necessary to study the behavior of these nanoaggregates under conditions that are representative for the gastrointestinal tract and the effects which may cause disaggregation (Ryzhakov et al. 2016).

Cyclodextrins as drug delivery vehicles

Although cyclodextrins can be found in 56 pharmaceutical products (source: Cyclolab), they are still regarded as novel pharmaceutical excipients, drug delivery vehicles, and anti-aggregation agents (Loftsson and Brewster 2010, 2012; Bilensoy 2011; Ahuja et al. 2011; Kurkov and Loftsson 2013; Chilajwar et al. 2014; Morin-Crini et al. 2015; Jansook et al. 2018). In the last decade, as already mentioned, it has been observed that cyclodextrins and cyclodextrin complexes in particular self-assemble to form nanoparticles and that, under certain conditions, these nanoparticles can self-assemble to form microparticles, a tendency that increases upon formation of inclusion complexes with lipophilic drugs. These properties have changed the way we perform cyclodextrin pharmaceutical research and have given rise to new cyclodextrin formulation opportunities as summarized by Bilensoy (2011), and Kurkov and Loftsson (2013). The design of functional cyclodextrin nanoparticles formed by self-assembly is also a developing area in the field of nanomedicine (cancer therapy) as reported by Fülöp et al. (2012). Nanoparticle-based systems can improve bioavailability, reduce immunogenicity, modify drug metabolism, reduce toxicity, and increase the biological half-life of drugs after systemic administration (Bilensoy 2011; Fülöp et al. 2012; Kurkov and Loftsson 2013; Tejashri et al. 2013; Concheiro and Alvarez-Lorenzo 2013; Simoes et al. 2014; Morin-Crini et al. 2015; Sharma and Baldi 2016).

Oliveri and Vecchio (2016) reviewed the use of cyclodextrins and their derivatives as anti-aggregation agents in a number of proteins such as insulin, prion protein, and amyloid-beta, and some multimeric enzymes. There are many diseases that are correlated to protein misfolding and amyloid formation processes affecting numerous organs and tissues. There are over 30 different amyloid proteins and

a number of corresponding diseases including Alzheimer's diseases the most common neurodegenerative disease. Treatment of these diseases is still a goal to reach, and many molecules including cyclodextrins were studied in this perspective.

Actually, cyclodextrins are also perceived as dream molecules for the development of applications in biomedicine and nanomedicine including nanovectorization, such as, for instance, nanoparticles for drug delivery, innovative biosensors for detection of biological targets, biorecognition events, molecular diagnosis and medical imaging, gene therapy or tissue engineering (Concheiro and Alvarez-Lorenzo 2013; Zhang and Ma 2013; Tan et al. 2014; Simoes et al. 2014; Dong et al. 2015; Wu et al. 2015b; Yuan and Zhang 2016).

Medical devices including catheters, prosthesis, vascular grafts, and bone implants can also benefit from surface grafting or thermofixation of cyclodextrins. This explains the recent increase in the number of research papers dealing with these topics (Concheiro and Alvarez-Lorenzo 2013). However, most of these studies are in the proof-of-concept stage, and only a few therapeutic nanosystems have been comprehensively investigated. The successful translation of these laboratory innovations to clinical reality remains challenging.

A new generation of drug eluting stents based on the strong anchorage of a biocompatible and bioresorbable cyclodextrin-based polymer onto metallic devices has been elaborated (Perez-Anes et al. 2015). Polydopamine, a strong adhesive polymer, was applied as a first coated layer. Cyclodextrin was fixed by in situ polycondensation with citric acid (polycyclodextrins formation). As a third layer an amine-rich polymer, polyethyleneimine, was used to stabilize the anionic cyclodextrin layer. As an alternative polycyclodextrins and chitosan layers were applied using layer-by-layer deposition technique.

Functionalized cyclodextrins are interesting scaffolds for contrast agents used in magnetic resonance imaging as reported by Gouhier's group (Idriss et al. 2013; Zgani et al. 2017). For the further improvement of the sensitivity of this medical diagnostic tool, it is necessary to fully understand the role of the cyclodextrins in the efficiency of the contrast agents. The review by Mavridis and Yannakopoulou (2015) can be also consulted on the same topic.

Cyclodextrins as active ingredients

Currently, the field of medicine is closely concerned with cyclodextrin inclusion complexes. These complexes are preformed prior to be administrated. There are, however, some cases when the complexes are formed within the body. The best-known example is the one containing the active compound sugammadex (Bridion®): it is a modified γ-CD used as an antidote to certain curare-like muscle relaxants in anesthesia since 2008 (Booij 2009; Yang and Keam 2009). After intravenous administration, it neutralizes steroid curare-like agents (rocuronium, vecuronium) by forming an inactive complex in the plasma which is then eliminated in the urine. Sugammadex has improved effectiveness compared with currently available methods of accelerating reversal of neuromuscular blockade. Its mechanism of action also differs from that of other commonly used reversal agents, e.g., neostigmine and edrophonium. Sugammadex is biologically inactive, does not bind to plasma proteins, and appears to be safe and well tolerated although it has few side effects. Its cost is markedly higher than that of any of the other drugs used in anesthesia (Donati 2011). The literature data are actually abundant with a matter of considerable debate. Sugammadex shows an unexpected rise from 17 papers in 2000–2005 to 780 in 2010–2015 (Source: Cyclodextrin News, Cyclolab).

Application of hydroxypropyl-β-cyclodextrin (HP-β-CD) against Niemann–Pick C disease (NPC) started with a surprising observation in 2007 that this cyclodextrin aimed as excipient was more effective than allopregnanolone, the active (Walkley et al. 2016). As no alternative treatment existed at that time the NPC1 disease families pushed FDA for approval for individual access for their children and as a result of a large collaborative work in all areas of drug development, including chemistry and manufacturing, formulation, pharmacology, pharmacokinetics, toxicology, and regulatory affairs, the preclinical clinical Phase 1 and Phase 2 studies have been concluded to start Phase 3 studies in 2015. In the meantime, the research focuses on the still not completely understood mechanism how HP-β-CD can help to NPC patients (Tanaka et al. 2015; Davidson et al. 2016).

Cyclodextrins are used in autophagy, a catabolic process with an essential function in the maintenance of cellular and tissue homeostasis. It is primarily recognized for its role in the degradation of dysfunctional proteins and unwanted organelles. Actually, the range of substrates also includes lipids. Autophagy is a self-degradative process that is important for balancing sources of energy at critical times in development and in response to nutrient stress. It also plays a housekeeping role in removing misfolded or aggregated proteins, clearing damaged organelles, such as mitochondria, endoplasmic reticulum and peroxisomes, as well as eliminating intracellular pathogens. Thus, autophagy is generally thought of as a survival mechanism, although its deregulation has been linked to non-apoptotic cell death (Glick et al. 2010). As cellular membranes play important role in autophagy, their modulation by cyclodextrins modifies this important housekeeping process of the cells. However, the results of various studies seem to be controversial partly because different cell types, different cyclodextrins were used at different concentrations. The

accumulation of autophagosomes, the intermediary products of autophagy, was interpreted both as a sign of activated and impaired autophagy. Hydroxypropyl-β-cyclodextrin treatment at high concentration leads to cholesterol depletion in an extent which leads to hindered fusion of the cellular membranes and this way to the diminished fusion of autophagosomes with lysosomes or to reduced expulsion of the autophagolysosomes. At low HP-β-CD concentrations, however, the autophagy is not perturbed or might be even improved. A combination of cholesterol removal by HP-β-CD and autophagy stimulation by rapamycin or carbamazepine seems to be a promising strategy in the treatment of impaired autophagy in lysosomal storage disorders, such as Niemann–Pick type C disease (Maetzel et al. 2014; Ward et al. 2016). Other interesting works have been published on cyclodextrin role (Yokoo et al. 2015; Tamura and Yui 2015; Motoyama et al. 2015, 2016; Manchon et al. 2016). Hydroxypropyl-β-cyclodextrin was found effective in inhibition of leukemic cell proliferation at various leukemic cell lines as recently reported by Arima's group (Yokoo et al. 2015). Their results demonstrated that hydroxypropyl-β-cyclodextrin was a potential anticancer agent in leukemia. Further studies are needed to understand the effect of cyclodextrins on autophagy and specially to learn how these effects can be utilized in the therapy of various illnesses including neurodegenerative diseases and cancers.

Cyclodextrins are also used for prevention and treatment of atherosclerosis as recently reported by Zimmer et al. (2016). These authors published a comprehensively study on the anti-atherosclerotic and anti-inflammatory effects of hydroxypropyl-β-cyclodextrin used as solubilizing agent to increase cholesterol solubility.

A recent review on the effect of cyclodextrins on blood brain barrier summarizes the findings of several research groups on various animal models of the central nervous system diseases (Vecsernyés et al. 2014). The CD-mediated cholesterol modulations change the action of various proteins in the membrane of epithelial cells in blood brain barrier. These proteins play significant role in pathogenesis of stroke, cerebral hypoxia and ischemia, Alzheimer, Parkinson and Huntington disease, epilepsia, central nervous system infections, and brain tumor. Extensive research is going on to translate these effects into therapy.

The alarming spread of bacterial resistance to traditional antibiotics has warranted the study of alternative antimicrobial agents. Quorum sensing is a chemical cell-to-cell communication mechanism utilized by bacteria to coordinate group behaviors and establish infections (Hirakawa and Tomita 2013; Morohoshi et al. 2013; Miller et al. 2015; Okano et al. 2016). It is known that cyclodextrins can interact with *N*-acyl-L-homoserine lactones (AHLs), the main signaling molecules for the bacterial cell-to-cell communication quorum sensing system. Many bacteria regulate their cooperative activities through releasing, sensing, and responding to small signaling molecules. This mechanism called quorum sensing makes possible for a population of bacteria to behave as a multi-cellular organism in host colonization, formation of biofilms, defense against competitors, and adaptation to changing environment. Quorum sensing is a new target for the development of antibiotic agents.

The concept of complexation of signal molecules (AHLs or peptides) by cyclodextrins does not aim to kill the bacteria but to control their growth and to decrease their virulence (Hirakawa and Tomita 2013). The development of novel strategies in the prevention and treatment of biofilm infections are expected in the next years. This will be useful not only in medicine but also in cosmetic, textile, and packaging fields (Brackman et al. 2016; Silva et al. 2016).

Cyclodextrins and nanotechnology

Cyclodextrins and their derivatives have been successfully employed to create novel nanomaterials, often called nanosponges (Bilensoy and Hincal 2009; Goyal et al. 2011; Davis and Higson 2011, Tejashri et al. 2013, Chilajwar et al. 2014; Trotta et al. 2014, 2015; Mavridis and Yannakopoulou 2015). A broad spectrum of cyclodextrin-containing materials with versatile supramolecular architectures such as nanoparticles, nanosponges, nanomicelles, and nanovesicles has been synthesized to assemble functional platforms. For pharmaceutical and biomedical applications (Bilensoy and Hincal 2009; Goyal et al. 2011), nanomaterials can be formulated as oral, parenteral, topical or inhalation dosage forms. These materials have also found applications in nanomedicine. Cyclodextrin-based nanosponges have been also developed as tool for the delivery of anticancer drugs, e.g., paclitaxel, doxorubicin, 5-fluorourcil, and tamoxifen, as reported by Trotta et al. (2014, 2015). These innovative materials can be considered as a challenging technology for the development of innovative formulations, suitable for various administration routes for anticancer drugs.

Cyclodextrins and foods

In recent years, the growth of the functional foods industry has increased research into new compounds with high added value for use in the fortification of traditional products (Martina et al. 2013; Lopez-Nicolas et al. 2014; Calo et al. 2015; Sharma and Baldi 2016; Kfoury et al. 2016; Fenyvesi and Szente 2016; Zarzycki et al. 2016). One of the most promising functional food groups is those enriched in antioxidant compounds of a lipophilic nature. In spite of the numerous advantages reported for such antioxidant molecules, they may also have disadvantages that impede their use in functional foods, although these problems may be avoided by the

use of encapsulant agents such as cyclodextrins (Fenyvesi et al. 2016).

An excellent review of the most recent studies on the complexes formed between several important types of antioxidant compounds and cyclodextrins was published by López-Nicolás et al. (2014). This comprehensive review focuses on the contradictory data reported in the literature concerning the antioxidant activity of the host/guest molecule complexes, the different complexation constants reported for identical complexes, the bioavailability of the antioxidant compound in the presence of cyclodextrins, and the recommendations concerning the use of natural or modified cyclodextrins. The authors also concluded that cyclodextrins will act as secondary antioxidants, enhancing the ability of traditional antioxidants to prevent enzymatic browning in different foods. Another interesting review on the encapsulation of antioxidants such as flavonoids and phenolic acids and their applications in food products, food supplements, and also packaging has been published by Zarzycki et al. (2016).

A general overview on the nanoencapsulation of flavors and aromas was also given in a chapter published by Fenyvesi and Szente (2016). Starting with the history dating back to the sixties of the last century, the advantages were summarized, the approval status of cyclodextrins in food was evaluated, and the methods of preparation and analysis were shortly outlined. Flavor complexes in food processing to reduce the loss in color, odor, and taste were revealed. Several examples were given to illustrate the advantages of nanoencapsulation of flavors in aroma preserving food packaging.

Cyclodextrins and cosmetics

Encapsulation techniques using cyclodextrins as (nano)-encapsulating agents are increasingly used not only by food, aroma, and pharmaceutical industries but also by cosmetic, fragrance and flavor industries, toiletry and personal care sectors for improving the efficiency of odorant and aroma substances, odor control in perfumes, masking unpleasant smells and tastes of some compounds, improving the physical and/or stability of essential oils and volatile compounds, stabilizing volatiles by reducing or eliminating any losses through evaporation, modifying the physicochemical and/or biological properties of the guest to afford a protective effect, or transforming of liquid compounds into crystalline form (Bilensoy 2011; Hougier and Kircik 2012; Kfoury et al. 2016; Zarzycki et al. 2016; Fenyvesi and Szente, 2016). The fragrance and flavor industry is a large and innovate sector of the chemical industry. Fragrance chemicals are added to consumer products such as personal care products, perfumes, deodorants, laundry detergents. Encapsulation techniques using cyclodextrins are increasingly used by this industry for protecting fragile molecules (eye-drop solutions) and improving the efficiency of odorant and aroma substances, but also to avoid the degradation of flavors by processing or, on storage, allowing the use of minor amounts of flavors. The use of these host compounds is also promising for various emerging fields such as aromatherapy and cosmeto-textiles—these new products also called 'cosmeceuticals' (Bilensoy 2011).

Cyclodextrins and textiles

Cyclodextrin molecules can be an ecofriendly alternative and used as finishing agents in textile applications. In this sector, their main characteristics are: ecofriendly nature, cost-effectiveness and ease of production at large-scale, physicochemical (e.g., inclusion complex-forming ability, solubilizing activity, chelating activity, slow release of fragrances) and biological (e.g., biocompatibility, biodegradability, drug carrier ability, insecticidal delivery) properties. These properties can be applied to different areas of applications such as deodorant (odor absorption, stabilization of active ingredients), fragrance/aroma, UV protection, water resistance, antimicrobial resistance, flame retardancy, and also insect repellent. In general, cyclodextrins are grafted in materials using binding and cross-linking agents. The comprehensive reviews of Islam et al. (2013) and Voncina and Vivod (2013), and the previous works by Hebeish and El-Hilw (2001), Martel et al. (2002), Romi et al. (2005), and Abdel-Halim et al. (2010) can be consulted. Cyclodextrin is considered as a promising reagent in textile finishing. Cosmeto-textiles also meet an increasing demand on the market and the cosmetic and textile industries are on the forefront of the research on this topic. These innovative materials are increasingly used not only in these sectors but also in pharmaceutical and medical industries, and in food packaging. Development of textiles with an antimicrobial activity can be also useful for water and air treatment.

Cyclodextrins and separation techniques

Although an important number of works have been published since the 1980s, the sector of cyclodextrin-based chromatography and electrophoresis continues to interest the scientific community (Hongdeng et al. 2011; Xiao et al. 2012; West 2014; Scriba 2016). The number of publications continues to grow not only in high performance liquid chromatography, ultra-high performance liquid chromatography, capillary chromatography, and gas chromatography, but also in supercritical fluid chromatography. Actually, liquid chromatography is considered as a green separation technique, as it avoids the use of organic mobile phase and is an ideal

alternative technique with fast and efficient separation for the preparation and separation of pure substances, e.g., enantioselective separation, chiral extraction).

Scriba (2016) published an interesting update review on the contributions to the understanding of the binding mechanism between chiral selectors and cyclodextrins in analytical enantioseparations dating between 2012 and early 2016. The author showed that many tools are available nowadays to study the mechanism of enantiorecognition including spectroscopic techniques (NMR) as well as molecular modeling for the visualization and analysis of the dynamics of the process. Selectors such as cyclodextrins appeared advantageous due to the much broader range of applications for structurally diverse analytes. Either random substituted cyclodextrins, which are mixtures of isomers with similar structure, or single isomer cyclodextrins are used. This high versatility makes cyclodextrins the first-choice selectors. The single isomers have the advantage of uniform structure, while the random substituted cyclodextrins often suffer from the batch-to-batch reproducibility (Li and Vigh 2004; Benkovics et al. 2016).

Cyclodextrins and catalysis

Cyclodextrins have long been known to be good contributors to the development of catalytic processes. They have been used for mass-transfer catalytic reactions and for the production of new catalysts mimicking enzymatic activity (Macaev and Boldescu 2015; Hong et al. 2015). The following classification for catalytic applications of cyclodextrins was proposed by Monflier's group (Hapiot et al. 2006, 2011, 2014): (1) to significantly increase the rate and selectivity of reactions catalyzed by water-soluble organometallic complexes; (2) to design new water-soluble ligands for aqueous organometallic catalysis; (3) to stabilize catalytically active noble metal nanoparticles in water; and (4) to facilitate reactions catalyzed by supported metals or metallic powder in water. Cyclodextrins were used in organic synthesis as promoters or catalysts of different reactions, as components of artificial enzymatic systems, as stabilizers for the nanodimensional metallic catalysts. A comprehensive collection of recent breakthroughs in aqueous cyclodextrin-assisted supramolecular catalysis can be found in the review by Hapiot et al. (2014).

Macaev and Boldescu (2015) published a state of the art of cyclodextrins in asymmetric and stereospecific synthesis. Three topics were summarized: (1) cyclodextrins' complexes with transition metals as asymmetric and stereospecific catalysts; (2) cyclodextrins' non-metallic derivatives as asymmetric and stereospecific catalysts; and (3) cyclodextrins promoting asymmetric and stereospecific catalysis in water. The authors concluded that cyclodextrins and their derivatives can be a feasible alternative to traditional catalysts in a variety of reactions.

Cyclodextrin-based supramolecular architectures

The state-of-the-art reviews of the design of complex macromolecular architectures based on cyclodextrin were presented by Schmidt et al. (2014) and Dong et al. (2015) and comprehensively discussed. In particular, by an elegant combination of dynamic/reversible structures with exceptional functions, functional supramolecular polymers are attracting increasing attention in various fields such as biomedical, e.g., gene transfection, protein delivery, bioimaging and diagnosis, tissue engineering, and biomimetic chemistry, and material science (polymer science, nanotechnology). Although extensive work has been done on cyclodextrin-based supramolecular architectures, future research needs to take into account their precise physicochemical characterization (Dong et al. 2011, 2015; Schmidt et al. 2014; Agostoni et al. 2015; Karim and Loh 2016; Gontero et al. 2017; Valetti et al. 2017). Indeed, the understanding and the design of supramolecular systems require a detailed characterization with respect to stoichiometry, affinity, structure, heterogeneity, and supramolecular dynamics.

Cyclodextrins and sugar-based surfactants

Carbohydrate-based surfactants are today an important class of amphiphilic compounds which play an important role with both fundamental and practical applications. The growing interest in such compounds is due to, inter alia, their preparation from renewable raw materials, their ready biodegradability and biocompatibility, as well as other more basic reasons of practical, economic, and environmental order. When complexed with cyclodextrins, carbohydrate-based surfactants considerably increase their performance and potential application range (Dodziuk 2006; Villalonga et al. 2007; Li et al. 2011). The formation of CD/surfactant host–guest compounds leads to an increase in the critical micelle concentration and in the solubility of surfactants. The use of these new systems is promising as reported by Valente and Söderman (2014).

Cyclodextrins and click chemistry

Click chemistry describes a family of modular, efficient, versatile, and reliable reactions which have acquired a pivotal role as one of the most useful synthetic tools for functionalization of cyclodextrins with a potentially broad range of applications (Faugeras et al. 2012; Dondoni and Marra 2012;

Schmidt et al. 2014; Hou et al. 2016). Cyclodextrins modified by the click reaction are building blocks of superstructures used for drug delivery systems (Zhou and Ritter 2010; Nielsen et al. 2010), for the modification of macromolecular surfaces (Celebioglu et al. 2014a), for the preparation of cyclodextrin dimers and trimers (Tungala et al. 2013), and for the generation of various glycoconjugates (glycopeptides, glycodendrimers, etc.) (Kushwaha et al. 2013). A tutorial review with the development in thiol-ene coupling for peptide glycosylation was published by Dondoni and Marra (2012). Cyclodextrins will play a very important role in all these new developments.

Cyclodextrins and agrochemistry

An interesting sector for cyclodextrins is agrochemistry (Ho et al. 2014; Garrido et al. 2014; Fernandes et al. 2014; Campos et al. 2015; Yusoff et al. 2016). Indeed, the efficacy of cyclodextrins for pesticide formulations has been evaluated. Cyclodextrins can effectively encapsulate or bind the pesticide's active ingredients in the material's matrix with a sustained release profile and slow mobility in soil (Yusoff et al. 2016). Cyclodextrin molecules form complexes with a wide variety of agricultural substances such as insecticides, fungicides, herbicides, repellents, pheromones, and growth regulators (Luca and Grigoriu 2007; Venturini et al. 2008). This is at the origin of many benefits: modification of the physicochemical properties of the included guest, i.e., physical state, stability, solubility, and bioavailability, stabilization against the effects of light or biochemical degradation, and reduction of volatility. All these benefits are interesting during the preparation of the commercial formulations. It is important to note that most of the pesticides-CD inclusion complexes studied in the literature have used β-CD because of its lower price, and most of the published papers related to pesticides are not directly practice-oriented (Garrido et al. 2014; Fernandes et al. 2014).

Electrospinning of functional nanofibers with cyclodextrins

Electrospinning is one of the most useful techniques for nanofibers production due to its versatility and cost-effectiveness. Electrospinning of functional nanofibers with cyclodextrin molecules was proposed, and an important contribution was made by Uyar's group on this topic (Celebioglu and Uyar 2012, 2013a, b; Kayaci and Uyar 2012, 2014; Kayaci et al. 2013a, b, c, 2014, 2015; Celebioglu et al. 2014a, b, 2016; Keskin et al. 2015a, b; Aytac et al. 2015, 2016a, b; Aytac and Uyar 2016, 2017; Senthamizhan et al. 2016). Functional nanofibers incorporating cyclodextrin were developed via electrospinning using different polymers and reactions (cross-linking, grafting, click chemistry). Cyclodextrin molecules were also proposed to produce nanofibers without using any polymer carrier because cyclodextrin can form aggregates via intermolecular hydrogen bonding in their concentrated solutions or polymeric structures through cross-linked reactions. Various applications were target of including functional materials for textile and medical textile, drug delivery, control release of antibacterials, packaging and food applications, complexation and microencapsulation (essential oils, antioxidants), and environmental purposes (e.g., filtration, purification and separation processes).

Cyclodextrins and remediation

In the last two decades, cyclodextrins as complexing agents have attracted considerable attention in environmental science in terms of removal of pollutants present in all environmental compartments, soils, air, waters and wastewaters, and sediments.

Soil flushing using cyclodextrin-based aqueous solutions was employed to solubilize pollutants. Cyclodextrin molecules are used as additives to enhance efficiencies and reduce the treatment time compared to the use of water alone or conventional surfactants. The review by Atteia et al. (2013) can be consulted on this topic.

Cyclodextrins found their role in the environmental risk management. Both risk assessment and risk reduction technologies may benefit from the CD's ability of forming inclusion complexes with the typical organic contaminants in soils as reported by Gruiz et al. (2011). The solubility enhancement of these compounds of usually poor water solubility can be utilized. Extraction of soils with aqueous solutions of well soluble cyclodextrin derivatives (HP-β-CD or RAMEB, randomly methylated-β-CD) gives information on the contaminant fraction easy to mobilize. As this fraction is the most available for the soil microbes, extracting the soil with HP-β-CD solution is considered as a measure of the bioavailable, biodegradable fraction. Nowadays, this method has become a part of the everyday protocol to predict the microbial bioavailability of organic pollutants of soils.

Cyclodextrin-based materials for water and wastewater treatment include cross-linked polymers and nanosponges, membranes, nanofibers, and functionalized systems such as polymers, silica, and organic resins. Morin-Crini and Crini (2013) reviewed the developments in the use of cross-linked cyclodextrin-based polymers as complexing polymeric matrices for pollutant removal by oriented-adsorption processes. The authors summarized the features of these polymers and how they were used in decontamination applications. Numerous interesting studies on the treatment of real effluents can be found in the literature (Nagy et al. 2014;

Khaoulani et al. 2015; Morin-Crini et al. 2015; Charles et al. 2016; Euvrard et al. 2016; Alsbaiee et al. 2016).

Imprinting technology makes possible to develop sorbents with extremely high specificity (Lay et al. 2016). Cyclodextrin-containing molecular imprinting polymers and cyclodextrins grafted to the surface of silica or other support via imprinting technology offer multiple binding sites with tunable surface properties for sorption of selected chemicals, e.g., perfluorinated compounds (Karoyo and Wilson 2015).

Nanofibers containing cyclodextrin molecules are another interesting material for pollutant removal. All the results published by Uyar's group demonstrated that these new materials may have high potential to be used as air filters for the removal of organic vapor wastes, adsorbents/filters for pollutant removal from aqueous solutions, and also drug delivery system.

There are several other interesting works for synthesizing membranes and nanofibers containing cyclodextrins (Meng et al. 2014; Xiao et al. 2014; Ghemati and Aliouche 2014; Zhao et al. 2015; Zhang et al. 2015; Wu et al. 2015a; Norena-Caro and Alvarez-Lainez 2016; Wei et al. 2016; Costoya et al. 2017). For example, Norena-Caro and Alvarez-Lainez (2016) proposed two different methods for the production of electrospun polyacrylonitrile (PAN) nanofibers containing cyclodextrin capable of capturing formaldehyde, a common indoor pollutant. The former comprised the addition of cyclodextrins to PAN/dimethyl sulfoxide solutions and the subsequent electrospinning of the mixture. The latter involved the cross-linking of cyclodextrins on electrospun PAN fibers by alkaline hydrolysis and esterification with citric acid. The results showed that both functionalized nanofibers might be used for indoor air purification. However, functionalized fibers obtained by addition of cyclodextrins were more effective for capturing formaldehyde than fibers obtained by cross-linking of cyclodextrins.

Cyclodextrin-functionalized silica networks used as adsorbents/filters for environmental applications has recently received a lot of attention as reported by Gibson (2014), Samiey et al. (2014), Lee and Park (2015), Dinker and Kulkarni (2015), Vunain et al. (2016), Mahmud and Wilson (2016), and Yamamoto and Kuroda (2016). Two main classes of materials have been proposed: cyclodextrin-functionalized silicas prepared through grafting reactions and cyclodextrin-silica hybrid systems prepared through sol–gel or self-assembly process. These materials could be applied in the elimination, enrichment and detection of environmental pollutants in air and water samples.

Actually, fundamental research is also focusing on cyclodextrin-based nanoparticles for environmental applications (Taka et al. 2017). Indeed, nanosponges have not only been explored for their pharmaceutical applications but for water purification and wastewater treatment (Landy et al. 2012; Tong and Chen 2013; Taka et al. 2017). This emerging technology of cyclodextrin-based nanosponges is expected to provide technical solutions to water treatment.

Conclusion

This review gives an overview of recent selected works on cyclodextrins used in various fields. The increasing number of publications on cyclodextrins shows that there is always an increasing interest in these molecules and their applications from academic and practical point of views. The emergence of cyclodextrins as active and smart molecules rather than complexing molecules in numerous cosmetic, textile, therapeutic, and biomedical products seems to be the next step in the development of cyclodextrin technology. Further studies, contributions, and industrial developments are expected in the near future in the following domains: catalysis, bacterial resistance, anticancer drugs (chemotherapy), magnetic resonance imaging, biotechnology, e.g., biotransformation, fermentation processing, enzyme models, peptide and protein delivery, material science, e.g., wrapping materials and packaging, cosmeto-textiles, agrochemistry, and sensor applications.

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