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Editorial

Hot Topics in 2022 and Future Perspectives of Macromolecular Science

Ana M. Díez-Pascual

Universidad de Alcalá, Facultad de Ciencias, Departamento de Química Analítica, Química Física e Ingeniería Química, Ctra. Madrid-Barcelona Km. 33.6, 28805 Alcalá de Henares, Madrid, Spain; am.diez@uah.es

In 1920, Hermann Staudinger discovered that macromolecules consist of long chains of covalently linked building blocks and subsequently published the first paper on polymerization. Since this article, millions of works have focused on macromolecules and their extraordinary impact on humanity [1]. Our world is incomprehensible without polymers. However, what does the future for macromolecular science entail?

From the very beginning, Staudinger acknowledged the value of publishing to encourage a scientific exchange within the emerging macromolecular research community. In this regard, *Macromol* is a young, open access journal that offers a sociable and active home for the macromolecular scientific community which is open to novel ideas, even those regarding public uncertainty and especially those on open speech. We believe that a knowledgeable interchange will lead to the development of novel directions for the area of macromolecular science as well as for our civilization. This is particularly apparent in our current period of global change in which the use of polymers is gradually becoming extensive and there is a reconsideration emerging of the common applications, where related technological changes will be required.

Throughout the year of 2022, various hot topics have been published in our journal. Starting from conventional synthetic polymers such as polytetrafluoroethylene (PTFE) [2], poly-methyl methacrylate (PMMA) [3] and its fluorinated derivatives [4], as well as copolymers [5,6], linear, low-density polyethylenes (LLDPE) [7,8], poly(hydro)silanes [9], poly(ethylene glycol) (PEG)-based soft elastomers [10], polyvinylidene fluoride (PVDF) [11], and poly(vinyl alcohol) (PVA) and its mixtures [12].

One of the hottest topics published in *Macromol* centered around the well-known polylactic acid (PLA), an aliphatic polyester that can be produced from agricultural resources such as corn and through a ring-opening polymerization of the lactides [13]. In this context, biocomposites comprising natural fibers and recycled PLA-based matrices are regarded as very interesting due to the combination of their good mechanical and physical properties as well as their sustainability [14]. The tendency towards the usage and development of eco-friendly raw materials will be increasingly predominant within the coming years since various regulations are encouraging this idea; for instance, in the automotive industry, it will be compulsory that 85% of plastic components of a vehicle arise from a recycling process. Thus, short fiber biocomposites based on PLA/polycarbonate (PC) blends derived from processing scraps and reinforced with cellulosic fibers are very promising. Additionally, flax-reinforced PLA biocomposites are ideal candidates as an alternative to conventional PP composites [15]. Mixtures of PLA and PP reinforced with inorganic nanotubes [16] and hydroxyapatite [17] have also been reported. The modification of PLA via the incorporation of boehmite alumina and thermochromic dye can be used to develop a biodegradable, novel shape-memory polymer composite (SMPC) [18].

Other macromolecules which have been widely investigated include polymers such as polyaniline (PANI) and polypyrrole (PPY) [19–22]. Since they have positive charges, they can bind to the negatively charged bacterial membrane and penetrate it, thus hindering bacterial activities. Thus, conducting polymers can form polymer composites with metal,

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metal oxides, and nanoscale carbon materials as novel types of antimicrobial agents [23]. The increase in reactive oxygen species (ROS) from the composites of polymers–metal nanoparticles has harmful effects and induces cell death. Among such ROS, the hydroxyl radical with one unpaired electron in the structure is most effective as it can oxidize any bacterial biomolecules, leading to cell death [24]. Future works should focus on the combination of conducting polymers and their composites with antibiotics, small peptides, and natural molecules with antimicrobial properties.

On the other hand, given the fossil fuel crisis and the reduction in finite resources, the synthesis of green polymers is becoming essential. The term "green" refers to materials which have a biological origin and/or are biodegradable and are produced via sustainable processes conducted under mild conditions which do not require the use of chemical catalysts, toxic solvents, or reagents [25]. In this regard, enzymatically synthesized bio-based and/or biodegradable polymers are gaining a lot of interest [26]. For instance, the enzymatic polymerization of PLA and alipharomatic furan-based polyesters (e.g., PBF) has been reported. Thus, significant actions have been taken toward synthesizing green polymers. Additionally, the biodegradation and chain scission of biodegradable polymers is of great interest. Thus, the epimerization and chain scission of PLA in the presence of various organic bases, including an alkylamine, a pyridine derivative, an amidine, and two phosphazenes, some of them being well-known organocatalysts for the ring-opening polymerization of lactide, has been investigated [27]. Additionally, the biodegradation of poly(ε-caprolactone) (PCL) loaded with 2 wt% $PbZrO_3$ has been examined [28]. The chemiluminescence results revealed that the degradation of PCL is accelerated in the presence of foreign atoms (Cr, Nd, Mg, Mn, and Ti) at the concentration of 0.1 mol%.

Numerous novel biopolymers and their composites have been investigated [29]. Natural biopolymers include cellulose, chitosan, starch, collagen, gelatin, hyaluronic acid, alginates, fibrin, and pectin, which are widely found in nature. These biopolymers have displayed many interesting properties, including biocompatibility, biodegradability, and antibacterial activity [30]. However, the development of novel methods to be used at an industrial level is required. For instance, low-substituted hydroxypropyl cellulose have been developed and were compressed at different compression loads to achieve a different tablet porosity. It was found that the particle size and the percentage of the hydroxypropyl content have a significant effect on the disintegration behavior of this biopolymer [31]. Additionally, high-molecular weight hypromellose and hydroxypropyl cellulose are widely known, extended-release polymers. High molecular-weight HPC GXF provided a better processability at low temperatures and an adequate tablet strength for the melt granulation of metformin HCl [32]. Other novel biopolymer blends such as Trigonella foenum–graceum galactomannan and xanthan gum mixtures have been prepared and their rheological properties have been assessed [33]. The synergistic interaction between two biopolymers, particularly in the samples treated with an ultrasound, resulted in better rheological aspects which could be related to the strong bonds between them.

Poly(n-alkylene succinate)s are a class of biodegradable and aliphatic polyesters that have attracted much interest, mainly in the framework of sustainability and the circular economy. Thus, the US Department of Energy (US DOE) declared biobased succinic acid as a chemical platform with a high potentiality towards the synthesis of compounds conventionally derived from fossil feedstock. During the last decade, succinic acid has been proven to be a green and sustainable precursor of many important, large-scale industrial chemicals and consumer manufactured goods [34]. Besides their green chemistry origin, these biopolymers are highly promising materials in a variety of sectors, including food packaging and biomedical applications. Their properties can be easily tuned, such as the semicrystalline morphology and the crystalline fraction, properties which are crucial for the applicability of these materials in several fields.

Biopolyamide 11/flax/basalt hybrid composites have also been developed to improve sustainability in the automotive sector [35]. Their impact performance as a function of temperature and plasticizer addition has been analyzed in detail. The results proved

that plasticized PA11 is endowed with a lower glass transition temperature and melting temperature, which makes for a simpler manufacturing and processing, but also possesses a higher toughness which delays the penetration phenomena and reduces the permanent indentation at room temperature.

Further, magnetic nanoparticle-reinforced biopolymers are also highly interesting for biomedical applications, including drug delivery systems guided by a magnetic field, image contrast agents, and heat generators in hyperthermia treatments [36,37]. Chitosan, a natural polysaccharide, has also been widely investigated as a biomaterial, especially for hemostasis. However, it presents some drawbacks, one of them being a low stability. Through chemical reactions with hydroxyl and amino groups, such as alkylation, carboxylation, quaternization, etc., different groups can be introduced into the repeating units and significantly enhance its hemostatic property [38]. It has also been used as a nanocarrier for drug delivery [39], as a scaffold [40], as antibacterial wound dressing [41] for food packaging [42], or for the treatment of illnesses [43]. The mixture of chitosan and PCL also shows great potential as scaffolds for tissue engineering [44]. Other natural polysaccharides such as kefiran, the primary structural component of kefir grains, which are the gelatinous irregular masses that form the symbiotic cultures of lactic and acetic acid bacteria, have been recently investigated [45]. It was used for the fabrication of cryogel films in the presence of plasticizers, such as glycerol and sorbitol. Varying the ratios of the polysaccharide/plasticizer system, different physical (the film thickness, moisture content, and solubility) and mechanical (the tensile strength and elongation at break) properties were attained.

Bio-based polyurethanes (PU) have also attracted a lot of interest in recent decades and have mostly overtaken petrochemical-based PU in terms of challenges such as solid pollution, economic effectiveness, and the availability of raw materials. Many kinds of available bio-renewable sources as precursors for the production of polyols and isocyanates have been explored for the development of "greener" PU materials; these bio-based polyurethanes have a noteworthy potential to be used as future PU products, for the replacement of petroleum-based polyurethanes, due to increasing concern about the environment as well as their relatively low cost and biodegradability [46]. Vegetable oils are an excellent raw material for the production of PU adhesives. The most important component of vegetable oil is triglycerides, which are glycerol esters with three long chains of fatty acids of a varying composition depending on the source of the oil. As triglycerides hydrolyze to form a range of fatty acids and glycerols, polyols, and isocyanates, raw materials for the synthesis of polyurethane can be derived from them. Various vegetable oils which can be utilized for PU synthesis are castor oil, palm oil, canola oil, soyabean oil, and jatropha oil [47–49].

Another hot topic is hydrogels, which are synthesized by the homo- and copolymerization of functionalized acrylamides [50]. The gels swell in aqueous solution, and some of them (e.g., poly(N-isopropylacrylamide (PNIPAM [51])) also swell in organic solvents of a low polarity (e.g., dichloromethane), making them amphiphilic materials. Nanocomposites can be prepared via dispersing nanomaterials (metallic nanoparticles, graphene, and carbon nanotubes) inside the gels. PNIPAM-based nanocomposites show a lower critical solution temperature (LCST) transition of the gel matrix, which can be reached by thermal heating or the absorption of electromagnetic radiation [52]. The characteristic properties (the swelling degree and rate, LCST, solute partition, mass transport, hydrophilicity, and biocompatibility) can be tuned by changing the functional groups in the copolymers and/or the other components in the nanocomposite. The nanocomposite's properties are used to produce technological applications, such as sensors, actuators, a controlled release, biological cell scaffolds and surfaces, antimicrobial, carriers of bioactive substances, and so forth [53]. For instance, xyloglucan (XG), a branched polysaccharide composed of a central backbone of D-glucose units linked by $\beta(1\rightarrow4)$-glycosidic bonds, decorated with D-xylose units through $\alpha(1\rightarrow6)$ glycosidic bonds, is a hydrogel-forming polymer which is able to retain large amounts of water. Its main applications are as a medical device for wound

dressings, mucosal protection, and ocular lubrication, as well as its uses as an excipient have been recently highlighted [54]. Additionally, new water-soluble photoinitiators based on the α-alkoxy aryl ketones have been synthesized and investigated for their ability to initiate photopolymerization for the preparation of hydrogels [55].

Overall, macromolecular science is always in need of a great diversity of competencies, from physical to biological and medicinal properties as well as from fundamental to applied topics. One recent challenge related with this growing interdisciplinarity is that an increasing number of polymer-related topics appear in novel scientific journals which are not directly associated with the macromolecular field. In addition to its interdisciplinarity, the macromolecular community is widely known and has been noticed by other chemical arenas for bridging academia with industry. The connexon between fundamental research and industry is crucial, as we have observed since the early days of polymer science.

A future world without polymers is unconceivable: they are currently used in many novel areas such as heat insulation, construction materials, microelectronics, green energy generation, soil fertility, food packaging and safety, the search for new antibiotics, regenerative medicine, ink-jet-based manufacturing, lightweight composite materials for windmills, and e-mobility, to mention but a few from an endless list.

The use of renewable resources for the synthesis of polymeric materials must be developed further because it offers the potential to replace petroleum-derived plastic materials. In terms of sustainability, this offers a noteworthy reduction in CO_2 emissions, displaying how polymers can play a role in reducing the use of fossil resources. The polymer industry and the related market will change in the coming future, but it continues to be productive and provide a range of opportunities.

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