



Light-induced release of molecules from polymers



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ABSTRACT

The release of molecules from polymers upon light stimulus has been investigated for a range of applications in particular for drug delivery. In this review, the concept of light-induced release processes from polymers is summarized. Light-triggered processes can be divided into two approaches, the light induced degradation of polymers and the light-induced polarity change of the polymers. Functional groups that can enable the breakdown of the polymer or the cleavage of a linker between polymer and small molecule encompass coumarine and *o*-nitrobenzyl groups while azobenzene and spiropyran undergo reversible changes. Although the literature is dominated by these four compound classes, functional groups such as anthracene, pyrene, perylene, 2-diazo-1,2-naphthoquinone, and BODIPY can undergo similar changes. Degradation of polymers or simple polarity changes can trigger the release of small molecules such as drugs, but also gas molecules such as nitric oxide and macromolecules including DNA and proteins can be liberated upon light-trigger.

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1. Introduction

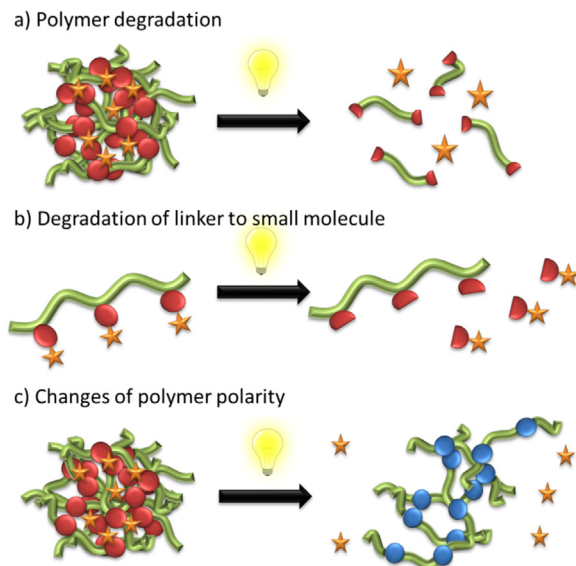
Light, as a source of energy to trigger chemical reactions, has attracted increasing attention in various fields including photopolymerization reactions and polymeric material design [1–26], 3D printing [27–39], light-induced ligation [40–44], organic synthesis [45,46], photocatalytic processes [47–50], and photodegradation of pollutants [51–54], to name but a few. Moreover, a range of parameters such as light intensity, emission wavelength, pulse length, and exposure time can be adjusted precisely to control reaction processes thanks to the availability of diverse light sources and their high degree of spatial and temporal resolution [55]. Among the applications, the combination of polymer-based drug delivery systems and light-triggered drug release processes is extremely appealing as it allows for spatiotemporal control of the photoresponse of polymeric systems in an accurate and robust fashion [3,55–76]. Specifically, well-designed polymer-based delivery systems can transport the molecules of interest (such as drugs) to the target sites within the body, followed by release of the molecules in response to external stimuli (e.g., change of temperature and pH values, magnetic fields, ultrasound vibrations, and light) [69,77–94]. Light-triggered release of drugs offers, compared to other stimuli, the advantage that it can operate in a spatiotemporal manner triggering the release of drugs at high local concentrations [57,59,75,95–105].

The design of photoresponsive polymer-based delivery systems relies on the introduction of photosensitive groups as part of the polymer structure that can absorb light to trigger photochemical reactions, which subsequently lead to the change of the properties of polymers triggering the release of loaded molecules. This is typically achieved either by triggering degradation of the polymer or a linking group or by introducing structural changes to the polymer that can cause a polarity shift (Scheme 1). This review summarizes the recent progress and development of polymer-based delivery systems for the release of small organic molecules, macromolecules, and gas molecules under various light irradiations.

2. Typical photoactive moieties used for light-induced release from polymers

2.1. *Ortho*-nitrobenzyl-based polymers

Ortho-nitrobenzyl derivatives are one of the most common photoactive compounds used in polymer-based drug delivery systems [106]. The mechanism of light-induced release process is thoroughly investigated and understood as illustrated in Scheme 2 [76,107–109]. Specifically, upon light irradiation, photochemical reactions via a series of reactive intermediates in the *ortho*-nitrobenzyl compounds and induce cleavage reactions take place with the result that the X moiety is released (Scheme 2). The cleavage of X can be used directly for the release of specific compounds from the polymer. Alternatively, the release can induce the change of polymer properties, which in return initiated the release of molecules.



Scheme 1. Typical polymer changes triggered by light enabling the release of molecules.

2.2. Coumarin-based polymers

Coumarin derivatives find applications in various fields including polymer science, medicine, and biology [110]. Coumarin can undergo dimerization upon light irradiation, a process that is reversible depending on the wavelength (Scheme 3) [111,112]. As a result, the properties of the polymers containing this moiety can be changed and the capsulated compounds thus can be released.

Coumarin compounds can be modified to make the light absorption red shift to blue region [113]. In addition, well-designed coumarin derivatives can also exhibit sufficiently high two-photon absorption cross-sections, making them attractive for photorelease applications in vivo [114].

2.3. Azobenzene-based polymers

Azobenzene can be isomerized (Scheme 4) on a timescale of microseconds or even sub-nanoseconds upon light irradiation reversibly for 10^5 – 10^6 times before fatigue [115]. When incorporated in polymeric materials, significant intra- and inter-molecular changes can be induced with suitable light irradiation due to fast, reversible, and innocuous *cis*-*trans* geometrical isomerization of azobenzene moieties [60]. Azobenzene-containing polymeric materials thus can find various applications in information storage, reversibly wettable surfaces, photomechanically responsive, biological and bio-mimetic systems, and drug delivery systems [60,116–119].

2.4. Spiropyran-based polymers

Colorless hydrophobic spiropyran derivatives can undergo photoinduced ring-opening reactions under UV light irradiation to produce their isomeric hydrophilic merocyanine forms, which

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