Conjugated Polymers and ROMP: Synthesis, Applications, and Prospects for Technological Innovations

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Conjugated Polymers and ROMP: Synthesis, Applications, and Prospects for Technological Innovations

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Abstract:

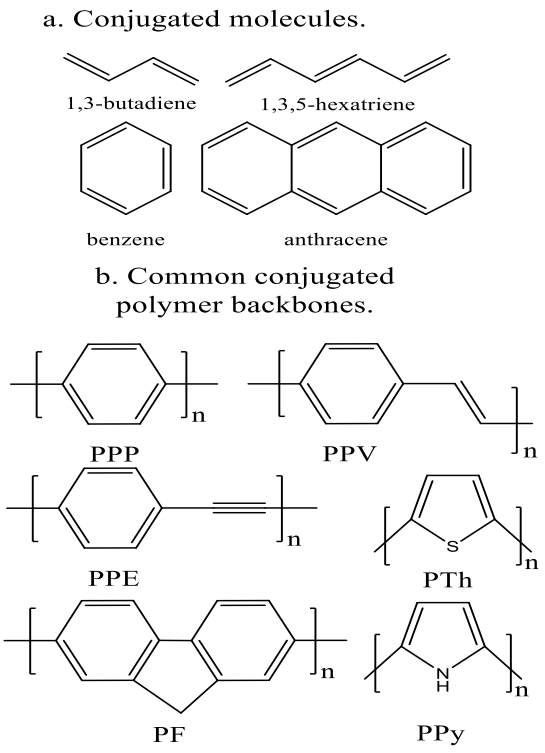
This article presents a succinct review of conjugated polymers, with special attention given to the significant impact of ROMP in their synthesis. ROMP enables precise control over the architecture of polymers and facilitates the integration of diverse monomers, Notably, PPVs and Norbornene derivatives have become commonly used ROMP monomers due to their high reactivity and compatibility with various functional groups. The review also explores recent developments in the practical application of conjugated semiconducting polymers. These studies demonstrate the promising potential of these materials across different industries and fields of research. With the versatility of ROMP, researchers can produce tailor-made materials with enhanced optoelectronic properties, opening up new possibilities for technological advancements. As research continues to progress, conjugated polymers are poised to revolutionize areas such as organic electronics, flexible devices, and bioelectronics, offering exciting prospects for future innovations.

Keywords: Conjugated polymers (CPs); Ring-opening metathesis polymerization (ROMP); Poly (p-phenylene vinylene) s (PPVs); Poly (norbornene dicarboximide) s (NDI); Grubbs catalyst (G).

Introduction

CPs are organic materials distinguished by a sequence of alternating σ and π bonds along their central structure. The appealing optical and electrochemical characteristics arise from the existence of a delocalized π-electron cloud along the backbone of these compounds [1–5].

In 1977, Alan MacDiarmid and Hideki Shirakawa [6] discovered and pioneered the conducting nature of CPs, a groundbreaking achievement that earned them the chemistry Nobel Prize in 2000. Since then, CPs have garnered substantial attention due to their promising technological potential. CPs can be obtained with different backbone chains, such as poly (p-phenylene vinylene) (PPV), polyfluorene (PF), polythiophene (PTh), poly(p-phenylene) (PPP), poly (p-phenylene ethynylene) (PPE), and polypyrrole (PPy). [5], etc. as shown in **Scheme. 1**. The fundamental feature of CPs can be described as a sequence of alternating double (or triple) and single bonds. This arrangement not only provides thermodynamic stability to the structure but also imparts specific properties, such as the capacity to absorb and emit light or conduct electrical charges. These distinctive characteristics make these materials highly versatile and valuable for diverse applications in chemical, biological, and material science fields.

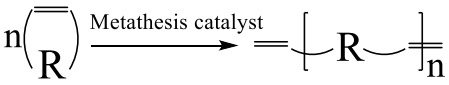


Scheme. 1 a) conjugated molecules; b) common conjugated polymer backbones

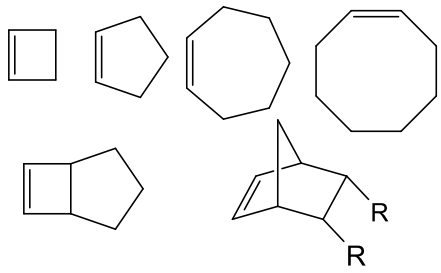
ROMP is a recently developed technique in the synthesis of CPs. Recognized as a highly influential controlled, and

living polymerization technique, ROMP enables the synthesis of well-defined polymers under mild reaction conditions **Scheme. 2**. The advantages of ROMP in polymer synthesis are numerous, encompassing superior selectivity, precise control over polymer architecture, and the capacity to integrate diverse monomers [7–12]. With this versatile approach, polymer structures and properties can be finely tuned, enabling the creation of materials with improved optoelectronic attributes.

a. Representative metathesis polymerizations.



b. Representative ROMP monomers.



Scheme. 2 a) representative metathesis polymerizations; b) representative ROMP monomers

ROMP monomers can consist of strained cyclic olefins that lack bulky groups around the double bonds. Norbornene and its derivatives are the preferred choice in ROMP due to their pronounced ring strain and ability to easily accommodate substituents in the ring structure. [12–14].

PPVs derivatives and NDI derivatives can be synthesized and incorporated into CPs using the ROMP method. The versatility of ROMP using the G catalyst allows the synthesis of various types of polymers; this includes the formation of both linear and cyclic polymers with substantial molecular weights and narrow molecular weight distributions. ROMP occurs through the opening of cyclic olefins, which are strained and can be activated by G catalysts to form highly reactive carbene intermediates that eventually undergo polymerization [3,4,14–19]. The present review is structured as follows: The initial section provides an introduction to CPs. The subsequent section concentrates on the synthesis of PPVs, NDI, and its derivatives. Lastly, we provide a summary of the recent applications of CPs.

1. Conjugated polymers

The identification of π-conjugated polymers and their distinctive optical and semiconducting characteristics has generated considerable attention in the domains of plastic electronics and photonics. The synthesis of these polymers with narrow optical bandgaps, tunable energy levels, and desired electronic properties has been particularly exciting due to their versatility of functionalization, thin film flexibility, low cost, and ease of processing. They have found comprehensive applications in organic field effect transistors (OFETs), organic photovoltaics (OPVs), fluorescent imaging, and organic light-emitting diodes (OLEDs) [5,20].

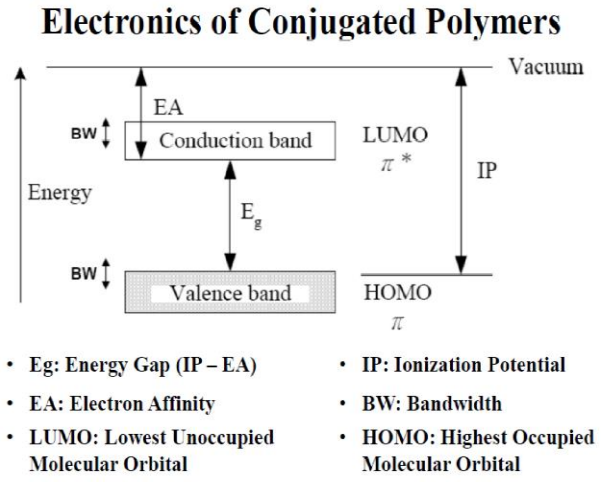


Figure 1. Electronics of conjugated polymers

CPs are comprised of a linear sequence of overlapping p_z orbitals exhibiting sp2 or sp hybridization. This results in a conjugated chain of delocalized electrons, which facilitates the movement of charge carriers within the backbone. The lower bandgap of the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) promotes the easier delocalization of π electrons, thereby enabling the injection of electrons into the LUMO and holes into the HOMO. To narrow the bandgap, various techniques have been employed, including the incorporation of electron-donating or electron-withdrawing groups into the conjugated backbone or the modification of the conjugated chain's length. **Figure 1**, When a photon is absorbed, it creates an exciton within the organic molecule or polymer. Subsequently, the excited electron transitions from the LUMO to the HOMO, releasing a photon during this process. This phenomenon is known as fluorescence. **Figure 2**. Research efforts have focused on the synthesis of donor-acceptor polymers. Aromatic benzene rings are commonly used to replace olefins, providing a large aromatic resonance stabilization energy and enhancing donor material efficiency [21–24].

The optical characteristics of CPs are defined by their bandgap, which is determined by the relative positions of the HOMO and LUMO with respect to vacuum. Spectroscopically, these bandgaps fall into distinct ranges: near-infrared (NIR) (1.59 to 0.4 eV), visible (VIS) (3.1 to 1.5 eV), and mid-infrared (MIR) (0.4 to 0.025 eV). Conjugated polymers commonly employed in organic solar cells and photodetectors possess NIR bandgaps,

while those utilized in light-emitting diodes exhibit VIS bandgaps. Further differentiation within the NIR region includes IR-A (1.6–0.9 eV) and IR-B (0.9–0.4 eV) ranges. In the past, low-bandgap polymers often exhibited shallow HOMO levels and were prone to chemical instability in their neutral and oxidized states, which imposed limitations on their practical utility. However, notable advancements emerged following the realization of polymer-based field-effect transistors, the achievement of electroluminescence in conjugated polymers, and the discovery of photo-induced charge transfer in conjugated polymer-fullerene blends. Consequently, chemists redirected their efforts towards synthesizing semiconducting polymers tailored for these specific applications. While LEDs benefited from semiconductors with bandgaps in the VIS range, the development of new low-bandgap polymers was undertaken to enhance the performance of organic photovoltaic devices.

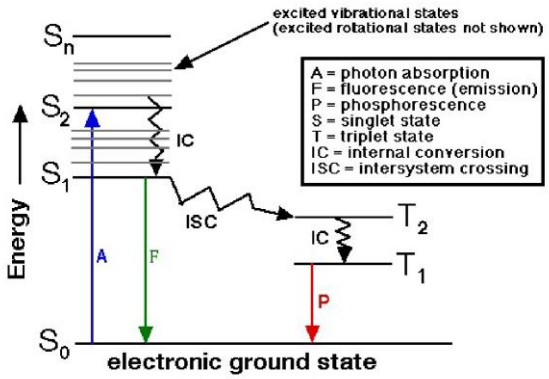


Figure 2. Jablonski diagram

In conclusion, the emergence of π-conjugated polymers with their remarkable optical and semiconducting properties has driven advances in the fields of plastic electronics and photonics. The ability to tune their energy levels, narrow their bandgaps, and exploit their charge carrier mobility has enabled diverse applications in organic photovoltaics and light-emitting diodes. Ongoing research continues to push the boundaries of their properties and applications, making them a promising avenue for future developments in the field of organic electronics.

2. ROMP Method

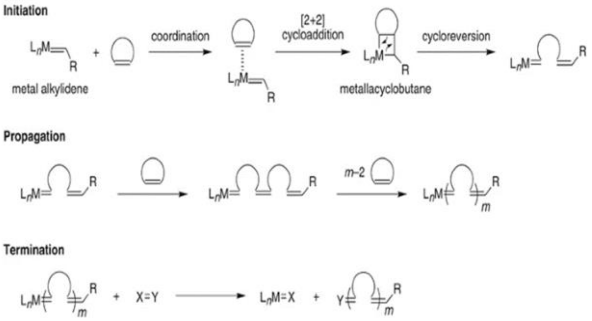
In the ROMP mechanism, the metal alkylidene complex must form a π-bond with the cyclic alkene, generating the metallacyclobutane intermediate. The bonds of this intermediate are then re-arranged by the breaking of the original double bond of the cyclic alkene and the formation of two new bonds, one between the alkene and the metal and the other between the alkene and alkylidene. Therefore, the original metal alkylidene bond is broken, and the next cyclic alkene is inserted into this bond. The structure remains tethered together by the original ring superstructure.

ROMP has three main stages **Scheme. 3**:

• Initiation: the formation of the reactive species

• Propagation: Numerous monomers are successively incorporated to create a polymer chain that retains the active terminal group.

• Termination: The process concludes when the active terminal group is neutralized through a reaction with a quenching molecule or due to the influence of moisture or oxygen, resulting in the formation of a polymer end group.



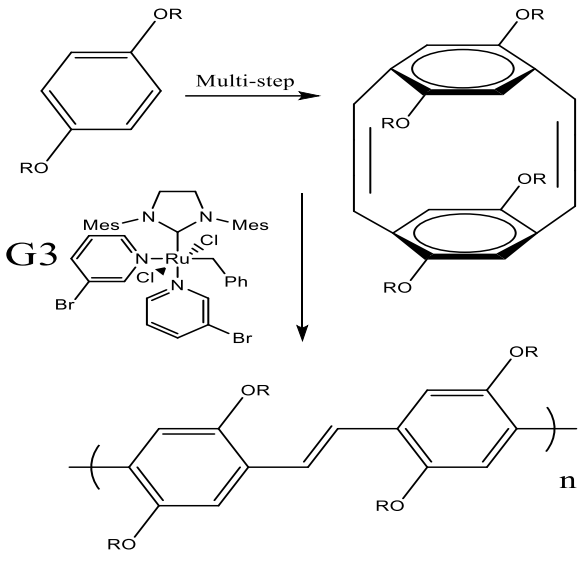
Scheme. 3 ROMP mechanism

Advantages of ROMP include the production of polymers of high molecular weight at low conversion (high yields) and complex block copolymer preparation. The limitations are that the monomers used must be cyclic and exhibit a high ring strain, especially for the ruthenium (Ru) catalysts [25].

3. Synthesis of PPVs, NDIs, and its derivatives

PPVs are CPs that have garnered extensive attention in both academic and industrial research. Nevertheless, many current manufacturing techniques lack meticulous command over essential factors like molar masses and alkene stereochemistry. These aspects play a pivotal role in influencing processability, polymer structure, and optical characteristics. In the conventional approach, the synthesis of PPVs relies on step-growth polymerization techniques such as palladium-catalyzed Stille, Suzuki, and direct arylation reactions. Although these methods yield a range of backbone structures, they encounter challenges related to inadequate regulation of molecular weight, wide disparities (Đ), and indistinct end groups.

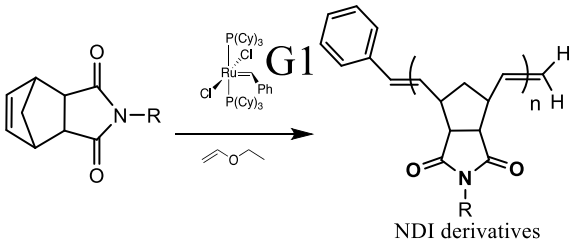
To address these shortcomings, novel synthetic strategies have emerged. These innovative approaches strive to generate PPVs with minimized defective polymer backbones, precisely controlled molecular weights, narrow dispersities (Đ), and clearly defined end groups. These advancements provide more precise and well-defined PPV structures, paving the way for improved properties and applications in various fields.



Scheme. 4 Polymerization of cyclophane-diene molecule

Alkoxy-substituted PPVs are easier to oxidize compared to the parent PPVs, resulting in significantly higher conductivities. Preparation of PPVs derivatives by the ROMP using Grubbs 3rd generation, starting withtetra-substituted paracyclophanediene **Scheme. 4**.

NDIs, which are extensively explored cyclic alkenes, are notable for their pronounced ring strain and the convenient attachment of dicarboximide units. This distinctive combination renders NDIs versatile for applications spanning various domains, encompassing chemistry, physics, and biology. In electro-optic (EO) applications like ring resonators, the solubility of organic constituents in common solvents is a critical requirement to facilitate processes such as spin coating. **Scheme. 5**.



Scheme. 5 Polymerization of NDIs

Moreover, the resulting thin films should exhibit minimal surface roughness, homogeneity, and superior optical quality. Interestingly, poly (NDI)s containing cycloalkanes not only fulfill these requirements but also demonstrate exceptional properties such as high glass transition (Tg) and thermal decomposition (Td) temperatures. These advantageous features make them highly attractive for various applications, offering a promising avenue for future advancements in relevant technologies.

4. Applications of Conjugated semiconducting polymers

4.1 Organic light-emitting diodes (OLEDs)

CPs have a wide range of uses in OLEDs, a unique technology that utilizes organic molecules to conduct significant charges, resulting in the emission of bright light ideal for displays and general lighting applications. The pioneering OLED, introduced by Tang and Vanslyke [26], utilized small molecule organic compounds with well-defined structures and fixed molecular weights, making them suitable for OLED applications [27].

To achieve successful implementation of OLEDs, it is essential for the materials and device structures to address the inherent high resistivity of organic compounds and ensure balanced charge injection from electrodes into the organic layers.

OLEDs consist of thin film devices that contain multiple organic layers sandwiched between two electrodes, converting electrical current into light through organic emitters **Figure 3**.

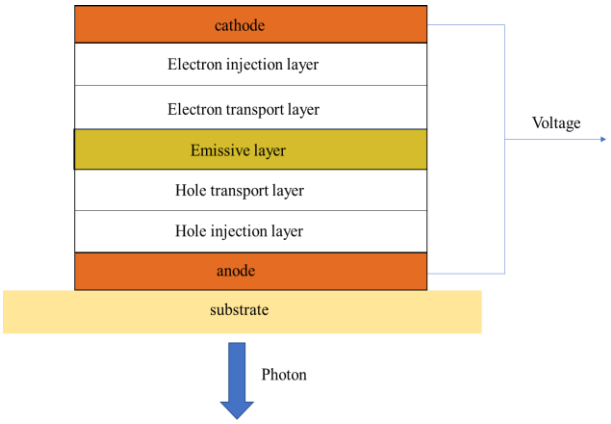


Figure 3. OLEDs design

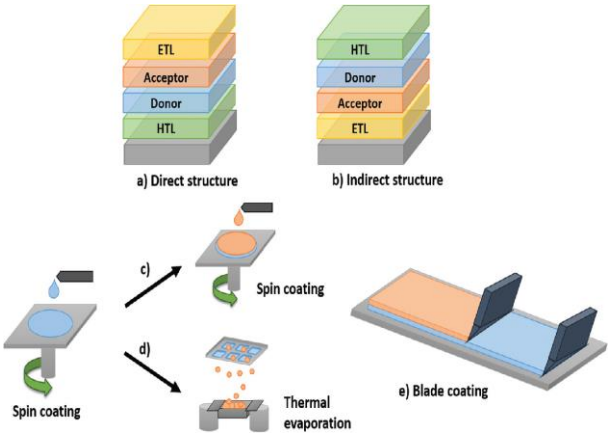
Organic compounds employed in OLEDs are carbon-rich and may include hydrogen, oxygen, nitrogen, or sulfur atoms. These materials, referred to as "soft," have relatively low melting points (usually below 350°C), allowing them to be compatible with flexible or nonplanar substrates like metal foils, plastic, and paper sheets. This flexibility enables the realization of innovative form factors such as lightweight, transparent, flexible, or rollable options.

In conclusion, the incorporation of CPs is instrumental in advancing OLED technology, offering the potential for efficient and adaptable light-emitting devices with diverse applications in display technologies and general lighting solutions.

4.2 Organic photovoltaic cells (OPVs)

CPs have opened up promising possibilities in the domain of Organic Photovoltaic Cells (OPVs). OPVs offer the potential for highly flexible, semi-transparent, and cost-effective solar cells, with an energy payback time of

several months. Despite initial difficulties, OPV cell efficiencies have made substantial strides in the last five years, now surpassing 18% and becoming a commercially viable technology [18,28]. To enable large-scale OPV production, critical factors include the selection of appropriate materials to ensure feasible mass manufacturing, avoiding low yields, and waste generation. Additionally, the fabrication and processing of active films play a crucial role in achieving consistent thin film structure and performance, necessitating adaptability to varying environmental conditions like humidity and temperature. Altogether, the applications of CPs in OPVs hold immense potential in advancing renewable energy technology, offering enhanced efficiency and flexibility for diverse practical applications **Scheme. 6**.



Scheme. 6 Diagram of a) a direct structure configuration; b) an indirect structure configuration of OPV devices and fabrication process via c) sequential spin coating; d) hybrid spin coating/thermal evaporation route; e) blade coating.

4.3 Organic field effect transistors (OFETs)

OFETs play a crucial role in the realm of electronic devices, featuring an organic semiconductor layer, a gate insulator layer, and three terminals (drain, source, and gate electrodes). Beyond their applications in cost-effective and flexible organic circuits, OFETs also offer valuable insights into the charge transport of π-conjugated systems. As a result, they serve as valuable tools for investigating the structure-property relationships of such systems, including parameters like field-effect mobility (μ), which represents the carrier drift velocity under an applied electric field; current on/off ratio, denoting the ratio of maximum on-state current to minimum off-state current; and threshold voltage, indicating the minimum gate voltage required to activate the transistor. Since their discovery in the 1980s, OFETs have attracted significant attention. leading to research on discovering, designing, and synthesizing π-conjugated systems tailored for OFETs [29]. The optimization of OFET devices and their applications covers a wide range, including uses such as flexible displays, radio frequency identification (RFID) tags, sensors, and electronic papers. **Figure 4** The present discourse concerns the configuration and constituents of bottom-gate top-contact thin-film transistors, in conjunction with the energy levels of the contact-semiconductor materials, wherein charge accumulation transpires.

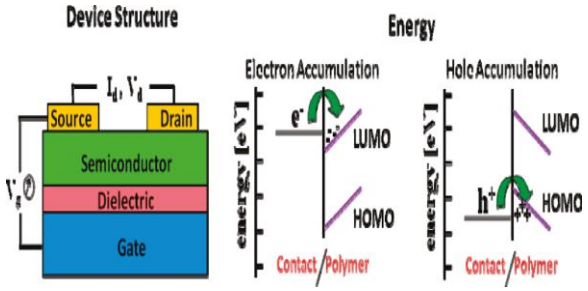


Figure 4. OFETs design

4.4 Fluorescent imaging agents

Fluorescence-based optical imaging has experienced remarkable development, allowing unprecedented spatial and temporal resolution to investigate various biological activities. However, the effectiveness of these advanced imaging methods heavily relies on the characteristics of the fluorophores used, as they influence detection limit, sensitivity, and reproducibility. Traditional organic fluorophores, while widely used, face challenges in high sensitivity cellular imaging due to their low intensity, tendency to photobleach, and rapid cellular clearance. Encapsulation in inorganic/polymeric nanocarriers can address some issues, but problems related to material and dye characteristics may lead to self-quenching, exclusion of the dye, and potential toxicity. Quantum dots, colloidal inorganic semiconductor nanoparticles, offer advantages like photobleaching resistance and narrow emission, but concerns remain about their degradation and cytotoxicity in vivo. Amidst these challenges, water-based luminescent nanoparticles formulated with conjugated polymers emerge as a promising class of fluorescent nanoprobes for bioimaging **Figure 5**. Their inherent conjugated backbone provides excellent optical properties, fluorescence brightness, photostability, rapid radiative rate, nonblinking behavior, and biocompatibility, making them an exciting alternative for in vitro and in vivo fluorescence probes [15,30]. Nonetheless, the quest for new and improved fluorescence probes continues, seeking to overcome existing limitations and advance biomedical imaging further.

4.5 Drug delivery systems

DDS based on polymers, which chemically encapsulate anticancer drugs through covalent linking to polymers, have garnered significant interest due to their biocompatible, biodegradable nature, and controlled drug release targeted to cancer cells or tumor tissues **Figure 6**. These CPs drugs have various advantages, including increased drug payloads, lower systemic toxicity, longer

drug circulation time, greater drug solubility, and improved targeting capabilities. Negatively charged poly (l-glutamic acid) has emerged as an ideal drug carrier, with various poly(l-glutamic acid)-drug conjugates being developed and utilized in clinical trials [31].

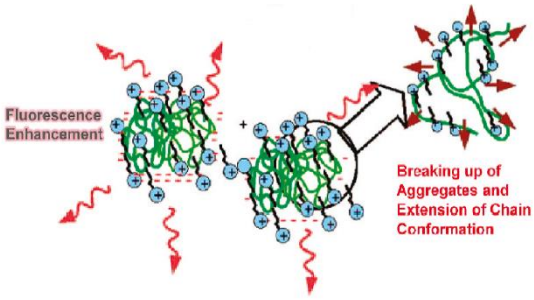


Figure 5. Proposed mode for the interactions between oppositely charged CPEs and surfactants at different concentrations

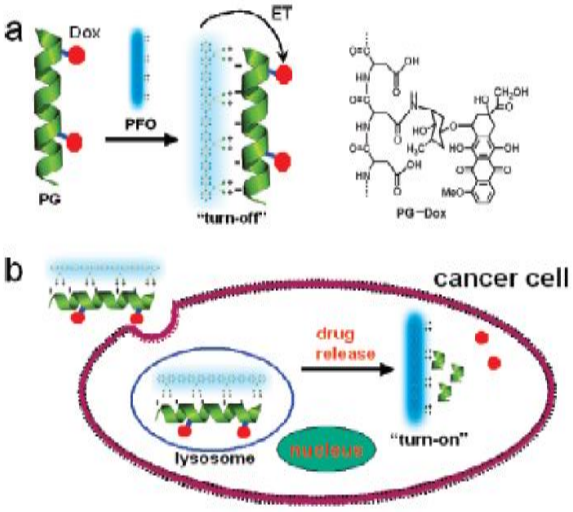


Figure 6. Example of DDS

Within the field of biomedical imaging, focus on multifunctional therapeutic systems. These systems seamlessly incorporate elements such as drugs, molecular targeting components, and imaging agents like fluorescent dyes and magnetic resonance imaging agents. This integration empowers noninvasive and real-time monitoring of drug delivery specifically to tumors, thereby bolstering the effectiveness of chemotherapy. Among these agents, fluorescent imaging agents hold particular significance in bioanalysis and imaging applications. As a result, Multifunctional polymer systems have gained widespread adoption as they allow for the accurate tracking of anticancer drug delivery and release. This is achieved by either loading or labeling polymeric carriers with suitable fluorescent imaging agents, thus facilitating precise observation of the entire process.

5. Conclusions

This mini-review emphasizes the introduction of CPs and the vital role of the ROMP method in their synthesis. ROMP provides precise control over polymer architecture and the incorporation of diverse monomers, enabling the creation of tailor-made materials with enhanced and tunable optoelectronic properties.

The versatility of ROMP with Grubbs catalysts allows for the synthesis of various CPs, including promising candidates like PPVs and NDI derivatives, known for their remarkable optical and semiconducting characteristics. CPs find applications in organic electronics, such as OLEDs and OPVs, offering efficient and adaptable light-emitting devices and cost-effective solar cells on flexible substrates. Moreover, their potential in OFETs and as fluorescent imaging agents demonstrates their value in affordable and flexible organic circuits and bio-imaging applications due to their optical properties and biocompatibility. In drug delivery systems, CPs show great potential for controlled and targeted therapeutic agent release, advancing personalized medicine and biomedical research.

Overall, the remarkable progress in CPs and the versatility of ROMP open doors for future innovations in organic electronics, flexible devices, bioelectronics, and biomedicine. Continued research holds exciting prospects for transformative solutions in various industries.

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