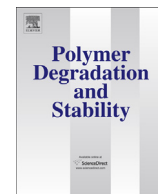




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High-tech functional polymers designed for applications in organic electronics

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ABSTRACT

Several examples of polymeric materials specially developed for optoelectronic applications are highlighted demonstrating that by fine-tuning chemistry and architecture in highly aromatic polymers significant progress can be made in their performance. Thus, polymeric semiconductors of controlled architectures and high charge mobility had been prepared. The use of branching allowed to realize highly aromatic polymers of high solubility suitable for solution processing as dielectric materials in OFETs and as light-outcoupling layer of high refractive index in OLEDs.

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1. Introduction

Traditional microelectronics is based on inorganic materials like silicon and silicon dioxide. These materials have limitations with respect to processability and flexibility. Polymeric materials can outweigh these drawbacks. Processability from melt and solution, high flexibility of films, light weight, and printability are reasons for a continuous and strong increase in interest in all-organic electronics. These properties open up the way to large scale optoelectronic applications via cost-efficient processes [1]. Moreover, the characteristics of polymers can be widely designed by their basic chemical structure. A variety of research results in the field of organic electronics has been reported so far [2–4] but only a few are presently used in practice. Possible applications for organic materials in electronic devices include among others organic light-emitting diodes (OLEDs) [5], organic field-effect transistors (OFETs) [6–8], organic photovoltaics (OPVs) [9], sensors [10], lasers [11], and radio-frequency identification (RFID) tags [12]. Requirements that polymers in general should meet for these applications are good availability, the opportunity to use solution-based deposition

processes at low temperature and their low-cost production [1,13].

In this contribution, we present some recent examples of polymeric material developments for organic electronic applications from our group. Here, we would like to emphasize, that besides the π -conjugated polymers, also polymeric dielectrics, as well as materials with special optoelectronic properties are highly important, since it is the combination of them that significantly determines the final performance of the device.

Thus, we will demonstrate the successful controlled preparation of naphthalenediimide [14] and dithienosilole (DTS) based conjugated polymers as well as block copolymers of DTS with thiophene [15] leading to promising semiconductor materials for printed OFET applications.

Furthermore, we will show that high-performance hyperbranched (hb) polymers with special optoelectronic properties are highly suitable for printable and polymer-based electronic applications due to their high processability as a result of the branching. E.g. hb polyphenylenes (hb-PPh) offer significant potential in the application as gate dielectric material which can be adopted for high performance OFETs. For multilayer device preparation, we also developed suitable crosslinking strategies which can be thermally or photochemically induced [16]. Furthermore, hyperbranched poly(vinylsulfide)s prepared through thiol-yne polyaddition [17] exhibited excellent high refractive index (HRI) properties and

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proved well-suited for the preparation of light out-coupling layers in OLEDs [18] and one-dimensional (1D) planar all-polymer photonic crystals (PhC) in organic lasers [19].

2. Chain-growth polymerization of unusual anion-radical monomers based on naphthalene diimide

Conjugated polymers are ideal candidates for printed organic electronics as they combine solution-processability with film-forming properties [20–24]. N-type (or electron-conducting) polymers are essential components in organic devices such as ambipolar and n-channel field-effect transistors or organic photovoltaics [24]. Strongly electron-deficient (n-type) main-chain π -conjugated polymers, e.g. based on naphthalene-diiimide [24,25] and perylenediimide [26,27] are commonly prepared via well-established step-growth poly-condensation protocols resulting in broad dispersities (approx. 2–5) and limited control over polymer molecular weight. Moreover, the often used Stille-coupling method utilizes highly toxic organostannyl derivatives. New synthetic protocols that allow high performance n-type conjugated polymers to be reproducibly prepared from non-toxic monomers via controlled polymerization methods are therefore highly desirable.

We reported a novel nickel-catalyzed chain-growth polymerization of an unusual anion-radical monomer based on the symmetric building block 2,6-bis(2-bromothiophen-5-yl)naphthalene-1,4,5,8-tetracarboxylic-N,N'-bis(2-octyldodecyl) diimide (Br-TNDIT-Br), polymerization of which leads to the corresponding bithiophene-naphthalene diimide conjugated polymer P(TNDIT) with low polydispersity and specific end-functions (Fig. 1) [14]. Our initial intention was to extend the scope of the chain-growth Kumada catalyst transfer polycondensation (KCTP) [28] towards electron-acceptor monomers. This requires the design and preparation of asymmetric AB-type monomers with a Grignard- and a halide-function in the same molecule. Such monomers are commonly prepared via magnesium-halogen exchange from the corresponding dihalide precursors and alkyl magnesium halides. Although electron-deficient aryl halides are usually more reactive in halogen-magnesium exchange reactions than electron-rich ones, we found that this method fails to give the Grignard monomer Br-TNDIT-MgBr upon the reaction of Br-TNDIT-Br with Grignard compounds. Another method to prepare monomers for the chain-growth polymerization of regioregular poly(3-alkylthiophene)s is Negishi polymerization (also referred to as "Rieke method") [29]. Especially its variation which utilizes activated Zn for the generation of organo-zinc monomers from aryl dihalides (i.e. Br-Ar-ZnX) is interesting here and was applied in order to generate polymers from Br-TNDIT-Br.

The polymerization of the Br-TNDIT-Br/Zn complex, which could be verified later on as an anion-radical (Fig. 1), was attempted next using various nickel complexes. Surprisingly enough, the addition of Ni(dppe)Br₂ or Ph-Ni(dppe)Br to Br-TNDIT-Br/Zn resulted in its rapid polymerization (a few hours until complete conversion) at room temperature. Detailed analysis of ¹H and ¹³C NMR spectra revealed a perfectly regioregular connection of the monomer units in the polymer structure via the 2-position of the thiophene rings. To clarify whether this polymerization involves a step-growth or chain-growth mechanism, several experiments were performed at different initiator-monomer ratios. It was found that the polymerization allows for a satisfactory control over molecular weight in a broad range of feed ratios (i.e. from 1/10 to 1/70 (Fig. 1) [14]. The successful polymerization with high conversion further corroborates the 1/1 Br-TNDIT-Br/Zn stoichiometry, since ZnBr₂ is formed as leaving group. As no insertion of Zn into the thiophene-bromine bond of Br-TNDIT-Br was observed, the discovered chain-growth polycondensation cannot be classified as Negishi/Rieke-type.

After additional studies and elucidation of the mechanism [14,30], we can state that a highly unusual Ni-catalyzed chain-growth polymerization of NDI-based anion-radical monomers leads to high-performance n-type conjugated polymers P(TNDIT) with controlled molecular weight, relatively narrow dispersity and specific end-functions. This result is outstanding taking into account the large length of the monomer (approx.1.5 nm) and its highly polarized structure. Intuitively, one could expect that both these factors hamper the intramolecular catalyst-transfer process, which obviously is not the case here.

The high performance of the n-type semiconductor P(TNDIT) could be demonstrated [31] in printed all-polymer OFET devices showing electron mobility values up to 0.2 cm² V⁻¹ s⁻¹ at a low work function of - 4.5 eV.

3. Dithienosilole-based all-conjugated block copolymers

Among the silicon-containing π -conjugated building blocks for conjugated polymers substituted dithieno[3,2-b:2',3'-d]silole (DTS) is extensively used as π -donor substructure due to its stability in ambient atmosphere [32–36]. Ohshita et al. [37] applied an anodic polymerization to a DTS-based monomer, however, this method suffers from low selectivity. As many other highly performing polymers, DTS-based homopolymers were synthesized by means of step-growth Stille coupling polycondensation, but this method leads to broadly distributed polymers which makes the preparation of high-quality block copolymers difficult.

In 2004/2005, McCullough et al. [38] and Yokozawa et al. [39] introduced an alternative chain-growth approach to conjugated

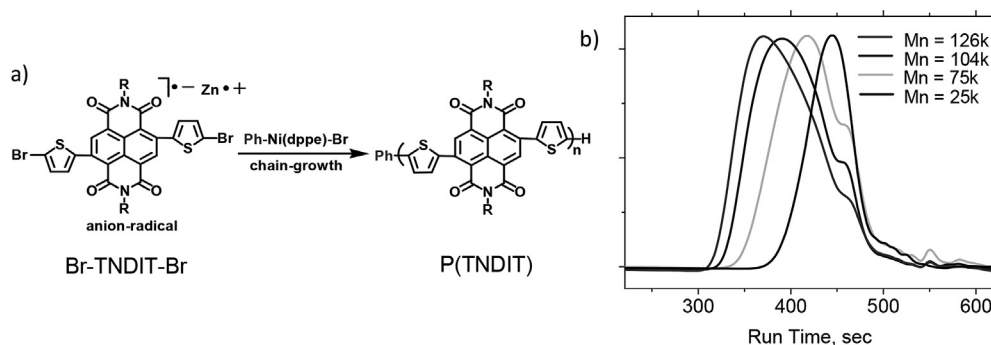


Fig. 1. a) Polymerization scheme towards P(TNDIT) from the Br-TNDIT-Br/Zn anion-radical and b) GPC elution curves of crude P(TNDIT)s in chloroform and the resulting weight average molecular weight (Mn) values (calibration with PS standards) obtained upon polymerization of Br-TNDIT-Br/Zn in the presence of Ni(dppe)Br₂ at the [initiator]/[monomer] ratios 1/10 1/30, 1/50, and 1/70 (from right to left) [adapted in part from Ref. [14]].

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