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Autonomous aqueous H₂O₂ production with
a carboxylate-functionalized polythiopheneCecilia Bruschi,^{†*ab} Asaminew Yerango Shimolo,^{†ac} Johanna Heimonen,^{ac}
Qilun Zhang,^d Mats Fahlman,^a Mikhail Vagin^{ab} and Renee Kroon^{ab*}

We report a novel carboxylate-functionalized polythiophene that spontaneously produces $\sim 1 \times 10^{-4}$ mol H₂O₂ per g polymer in the presence of water and oxygen, corresponding to $\sim 18\%$ polymer doping level. We show that this is a reversible redox process that allows for regeneration of the polar polythiophene. These results could open new applications that are driven by the autonomous production of H₂O₂ by conjugated polymers and elucidate degradation of similar CPs that are exposed to water and oxygen.

^aLaboratory of Organic Electronics, Department of Science and Technology, Linköping University, Bredgatan 33, 60221, Sweden. E-mail: cecilia.bruschi@liu.se; renee.kroon@liu.se

^bWallenberg Initiative Materials Science for Sustainability, Linköping University, Bredgatan 33, 60221, Sweden

^cWallenberg Wood Science Center, Linköping University, Bredgatan 33, 60221, Sweden

^dDepartment of Materials Science and Engineering, Stanford University, 496 Lomita Mall, Stanford, CA 94305, USA

[†] Cecilia Bruschi and Asaminew Yerango Shimolo contributed equally to this work.



Renee Kroon

His research is curiosity-driven, with a vision to combine green synthesis of conjugated polymers with stimuli-responsive conjugated polymer designs to create and investigate complex electro- and photoactive hybrid materials.

Renee Kroon is an Associate Professor of Organic Chemistry at the Laboratory of Organic Electronics, Linköping University since 2020. He received his MSc in Polymer Science in 2004 from the University of Groningen and his PhD in Polymer Chemistry in 2013 from Chalmers University of Technology under the supervision of Mats Andersson. He completed postdoctoral research working on functionalized conjugated polymers for thermoelectrics

Introduction

Low ionization energy (IE) conjugated polymers (CPs) are a class of materials that find prominent use in a myriad of organic electronic (OE) applications that are operated through aqueous electrochemical processes such as energy storage devices,¹ ion pumps,² organic electrochemical transistors (OECTs),³ thermoelectrics,^{4,5} charge-transfer doping⁶ and H₂O₂-production *via* electrocatalysis.⁷ Operation of all these devices involves the exposure of the CP to water (H₂O) and most often, unless actively removed or obstructed by barriers, atmospheric oxygen (O₂); an abundant and renewable oxidant whose presence plays a vital role in advancing sustainable chemical and energy technologies.

For example, in the electrocatalytic production of H₂O₂ with CPs, the presence of both O₂ and H₂O is desired to perform the oxygen reduction reaction (ORR). ORR proceeds *via* either a 4-electron pathway yielding water or a 2-electron pathway producing hydrogen peroxide (H₂O₂). Another proposed mechanism describes a 1-electron reduction process that, depending on the pH, forms either the superoxide radical (O₂^{•−}) or the hydroperoxyl radical (HO₂), which then undergoes disproportionation and protonation to H₂O₂ and O₂.⁸

In contrast, the formation of H₂O₂ in OECTs constitutes an undesired reaction since it would result in material degradation and compromise the lifetime of the device. The formation of H₂O₂ under neutral conditions was reported as an ORR product in OECTs based on poly(ethylenedioxythiophene):(polystyrene sulfonate) (PEDOT:PSS) or P(g₄2T-TT),^{9,10} indeed resulting in gradual degradation accompanied by a decrease in device performance and, thus, limiting the applicability of low IECPs in organic electronics. In the case of PEDOT:PSS-based depletion-mode OECTs, the H₂O₂ is produced through ORR only in their electrochemically reduced 'OFF'-state. Conversely, accumulation-mode devices based on neutral, low IE CPs such as P(g₂T-TT), are oxidized by O₂ at zero potential, but only when exposed to acidic media (pH $\approx$ 2.5).¹¹ In this case H₂O₂ has been proposed as one of the possible products of the ORR



driven on polymer, but no experiments were done to verify the formation of H_2O_2 .

An unexplored concept for H_2O_2 production with CP is the use of the virgin electron-rich CP that serves as a donor of electrons with sufficient capacity and energy to drive selectively ORR in a self-powered mode, a process akin to autoxidation, without the need for additional reagents. While autoxidation of organic molecules to H_2O_2 is already a known process for molecules like ascorbic acid^{12,13} and phenols,^{14,15} these reactions require the presence of metals to accelerate the autoxidation process^{16–18} or the presence of specific conditions such as pH and temperature.¹⁹ In addition, ascorbic acid is consumed in the process of autoxidation, forming the unstable dehydroascorbic acid (DHA) that undergoes irreversible hydrolysis to 2,3-diketo-L-gulonate.¹² The autoxidation of phenols leads to the formation of quinones, which require catalytic hydrogenation to regenerate its H_2O_2 producing properties. To enable the autonomous production of H_2O_2 with CP, the chemical design of the CP must allow for efficient unison of O_2 as the oxidant, H_2O as the proton source while acting as an efficient electron source itself. To achieve this, a low IE, hydrophilic CP design should be targeted, where the low IE offers a strong driving force for ORR while hydrophilicity increases accessibility of the CP to O_2 and H_2O for this reaction to occur.

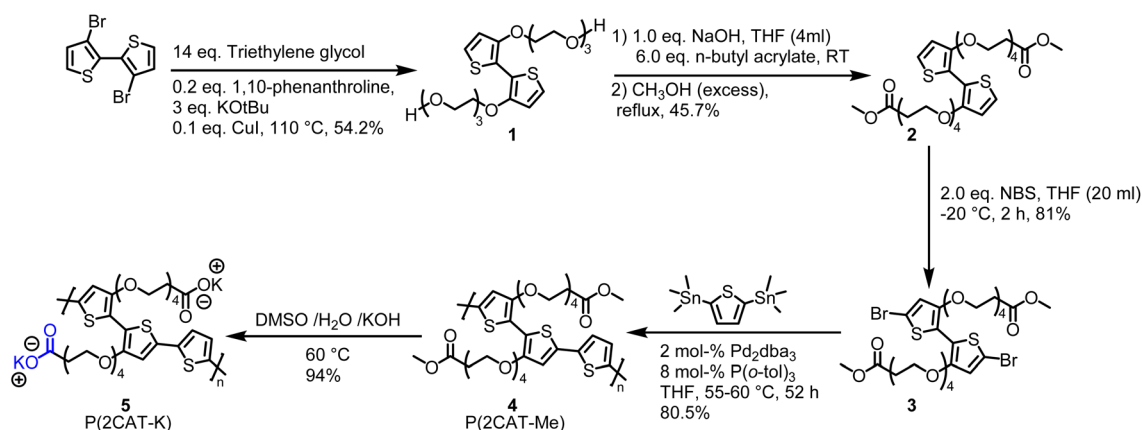
We demonstrate such a concept with a novel, low IE, carboxylate-functionalized CP (P(2CAT-K)), under neutral aqueous conditions. We present the synthesis of P(2CAT-K) with a low IE⁰ ~ -4.4 eV that exists as a nanodispersion in water. Exposure of aqueous P(2CAT-K) dispersions to O_2 triggers the slow, autonomous generation of H_2O_2 , without the need for external circuitry, which we monitored by changes in the polymer's UV-Vis spectra during the ORR process. We show that aqueous nanodispersions of P(2CAT-K), when exposed to O_2 , result in the autonomous formation of $\sim 1 \times 10^{-4}$ mol H_2O_2 per g of P(2CAT-K), which corresponds to a $\sim 18\%$ doping level of the polymer, and demonstrate that this process can be regenerated. We believe that the considerable amount of H_2O_2 produced by P(2CAT-K) could allow for new applications that are driven by autonomous H_2O_2 production of CPs. In addition, the autonomous H_2O_2 production by P(2CAT-K) can be extended to other low IE polymers that are exposed to oxygen and water and shed light on degradation

mechanisms in organic electronic devices that are operated under ambient conditions.

Results and discussion

The synthesis of the water-processable, carboxylate-functionalized polar polythiophene P(2CAT-K) (Scheme 1) starts with an Ullman coupling of triethylene glycol to 3,3'-dibromo-2,2'-bithiophene under neat conditions to afford compound (1). Compound (1) was then reacted with *n*-butyl acrylate *via* oxa-Michael addition, directly followed by *trans*-esterification with methanol to the methyl ester (2). Bromination with *N*-bromo succinimide then yielded monomer (3), which was finally polymerized with bis(2,5-trimethylstannyl) thiophene under Stille conditions to produce the precursor polymer, P(2CAT-Me) (4). Hydrolysis of P(2CAT-Me) with potassium hydroxide in DMSO:H₂O resulted in the water-processable P(2CAT-K). The structures of the intermediates, monomer, and polymers were confirmed by ¹HNMR and ¹³CNMR spectra. Detailed experimental procedures, ¹H and ¹³CNMR of monomer (S1–S6), polymers (S7 and S8), GPC data (S9) FTIR spectra (S10) can be found in the SI. Upon ¹H NMR analysis of the P(2CAT-K) in D₂O, we no longer observe peaks in the NMR spectrum, which suggests the formation of small particles rather than a true aqueous solution. To investigate whether P(2CAT-K) forms a dispersion, we performed dynamic light scattering (DLS). DLS analysis of a 0.3 mg mL^{−1} aqueous solution showed particles with an average size of 214 nm and PDI (Potential Distribution Index) of 0.7 (see Table S11 and Fig. S12). This result suggests that P(2CAT-K) exists as a nanodispersion in aqueous media rather than a true solution. As expected, decreasing the polymer concentration to 0.03 mg mL^{−1} and to 0.003 mg mL^{−1} leads to the detection of particles with a lower size of 139 and 133 nm respectively (Table S11). The formation of aggregates appears to be an effect of molecular weight, since lower molecular weight P(2CAT-K) displays peaks in the ¹H NMR spectrum (Fig. S8).

To investigate the redox properties of P(2CAT-K), electrochemical characterizations were carried out. Attempts to perform cyclic voltammetry (CV) of the polymer aqueous



Scheme 1 Synthesis route for the carboxylate-functionalized P(2CAT-K) polymer.



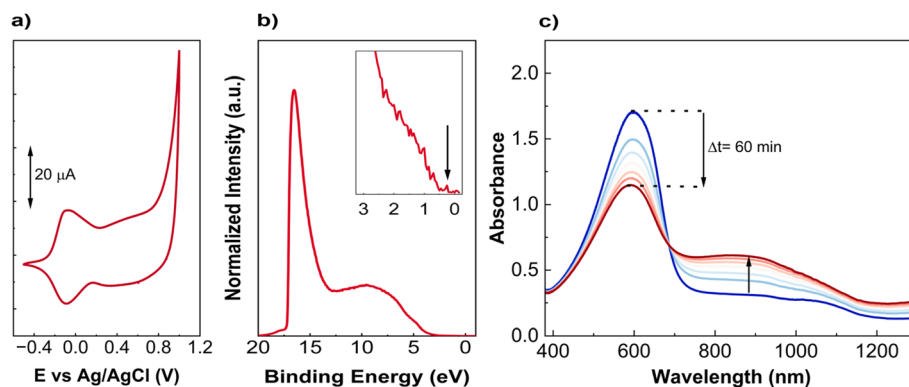


Fig. 1 (a) CV of P(2CAT-K) film (0.1 M KCl electrolyte, 5 mV s^{-1}); (b) UPS spectrum of P(2CAT-K) (thin film, iridium substrate); (c) evolution of the UV-Vis spectrum of an aqueous P(2CAT-K) nanodispersion in air ($\Delta t = 60 \text{ min}$, 10 min between scans).

dispersion failed due to precipitation of the polymer in the highly concentrated electrolyte solution, which we explain by a salting-out effect. Therefore, CV analysis was performed on a drop-cast polymer film deposited on a glassy carbon working electrode (Fig. S14) in KCl (0.1 M) electrolyte solution. The recorded CV (Fig. 1a) shows a switching region, correlated to the oxidative doping process, in the range from -0.3 to 0.2 V , followed by a conductive region, characterized predominantly by a capacitive current. The observed low oxidation potential suggests a low IE for P(2CAT-K). Attempts to perform a standard evaluation of ORR using hydrodynamic voltammetry, namely rotating disk ring electrode modified by P(2CAT-K) film, failed due to the high ORR activity of the current collector. To establish a value for the ionization energy, we performed ultraviolet photoelectron spectroscopy on a thin film of P(2CAT-K) and measured an IE^0 of $\sim -4.4 \text{ eV}$ (Fig. 1b), which is similar to the non-functionalized, low IE polar polythiophenes that were previously reported.²⁰

The solid-state absorption spectrum of the P(2CAT-K) (Fig. S16) was recorded in the visible and NIR region, after annealing the film at 100°C under N_2 to remove the doping induced by oxygen adsorption.²¹ The UV-Vis spectrum shows a parent absorption peak at $\lambda = 590 \text{ nm}$, similar to previously reported glycolated polythiophene analogues²² and to the one recorded for the aqueous polymer nanodispersion. We explain the minimal difference between the solution and solid-state absorption of P(2CAT-K) due to the formation of nanoparticles, which likely involves interchain aggregation. The absorption spectrum of a freshly prepared water dispersion of the polymer (see Fig. 1c) also shows the emergence of a broad absorption peak ($\lambda \sim 880 \text{ nm}$) covering mostly the NIR region, which is attributed to a polaron peak.²³ We observe that, after leaving the polymer dispersion for one hour under ambient conditions, the polaron peak shows a significant increase in intensity, while the polymer peak in the visible region decreases (Fig. 1c).

The appearance of the polaron peak without the addition of a chemical dopant, led us to hypothesize that the P(2CAT-K) is indeed oxidized by O_2 that is present in the solution with consequential formation of H_2O_2 through an ORR reaction. To exclude a photocatalytic process, we performed doping tests

with and without light exposure. We observe that the polaron peak intensity of both reactions, and thus the doping level, is of very similar intensity (Fig. S17). Therefore, we conclude that light is not a factor in the observed ORR and instead, there is a thermodynamic driving force for H_2O_2 production. To check the effect of aggregation, we also measured the ORR reaction for the low molecular weight, dissolved P(2CAT-K) and observed the same change in the UV-Vis spectra as for the nanodispersed P(2CAT-K) (Fig. S18).

Next, we developed experiments with larger amounts of P(2CAT-K) to be able to qualitatively and quantitatively analyze H_2O_2 . Aqueous nanodispersions of P(2CAT-K) (0.5 mg mL^{-1}) were prepared under inert conditions, which were exposed to different reaction conditions: one dispersion was sparged with O_2 , a second with N_2 , and a third left under ambient air. After one hour, an aqueous solution of CaCl_2 was added to each vial to precipitate the polymer through ionic crosslinking with Ca^{2+} , so that the aqueous solution containing H_2O_2 could be conveniently retrieved through filtration. The filtrated solutions were then analyzed on H_2O_2 concentration *via* the horseradish peroxidase/3,3',5,5'-tetramethylbenzidine (HRP/TMB) assay.^{24,25} The absorption spectrum of the solution purged with O_2 , once mixed with the HRP/TMB assay, exhibited the characteristic peak ($\lambda = 653 \text{ nm}$) of oxidized TMB, confirming the production of H_2O_2 *via* polymer oxidation by O_2 (see Fig. S19). An absorption peak with lower intensity at the same wavelength was observed for the aqueous nanodispersions exposed to ambient air, while an almost negligible peak was recorded for the dispersions sparged with N_2 (Fig. S19). The difference in H_2O_2 concentration is easily observable as a color intensification that progresses from the almost transparent solution sparged with N_2 to the strongly blue colored solution sparged with O_2 (Fig. 2b, inset). The amount of H_2O_2 quantified for the aqueous nanodispersions of P(2CAT-K) (0.5 mg mL^{-1}) sparged with O_2 was equal to $58.86 \pm 16.10 \mu\text{M}$. The finiteness of the number of electrons available in CP for ORR defines the linear dependence of oxidation level of P(2CAT-K) on the amount of produced H_2O_2 . Assuming 100% selectivity of 2-electron ORR to H_2O_2 , which is justified since several studies have concluded that the four-electron reduction of O_2 to H_2O does not proceed efficiently without combination with suitable inorganic



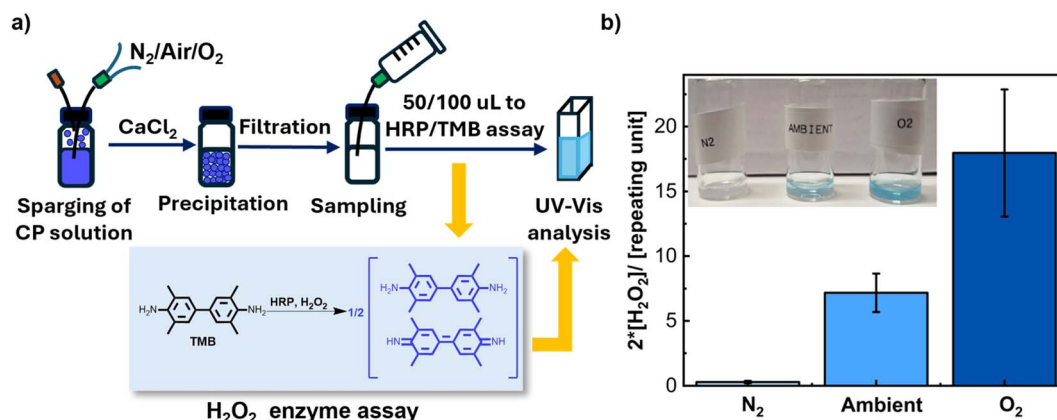
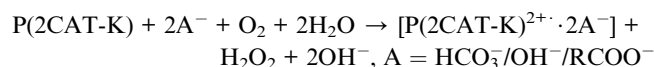


Fig. 2 (a) Scheme of the experimental procedure used for the autonomous production and quantification of H_2O_2 by P(2CAT-K); (b) oxidation level of P(2CAT-K) based on measured H_2O_2 concentration (multiplied by two) per number of polymer repeating unit obtained after exposing the same P(2CAT-K) aqueous nanodispersion to three different environments: O_2 , ambient air and N_2 atmosphere. Inset: Color of the filtrated solutions after addition to the HRP/TMB assay. The blue color is related to the charge-transfer complex formed by TMB in the presence of HRP enzyme and H_2O_2 .

cocatalysts,^{7,26–28} one can calculate the maximum oxidation level of the polymer using the known amount of moles of repeating unit (Fig. 2b). The maximum oxidation level of P(2CAT-K) obtained in oxygen-saturated system is then equal to 17.96 ± 4.91 which corresponds to an average amount of $\sim 1 \times 10^{-4}$ mol H_2O_2 per g P(2CAT-K). We explain the relatively high standard deviation because the total dataset includes experiments involving different solutions prepared and tested on different days, while we observed good reproducibility and small deviations in the results obtained within the same experimental set. Furthermore, we acknowledge that H_2O_2 production is solely due to kinetics. To evaluate the subsequent, more thermodynamically favoured reductions of oxygen and H_2O_2 to water, we ran ORR polymer-mediated experiments in the presence of added H_2O_2 , and we still observed the generation of H_2O_2 (Table S22). These results prove the polymer kinetically favours the oxygen reduction reaction to H_2O_2 .

Lastly, based on our results and previous literature studies, we propose a reaction scheme for the observed autonomous

H_2O_2 production using P(2CAT-K). The first step involves an outer-sphere electron transfer^{8,29} from the polymer to dioxygen. The potential inversion provides a significant driving force for the second electron transfer from the polymer, which then occurs immediately. Both electron transfers result in the formation of polarons in the polymer backbone. We propose that the formed positive polaron could be stabilized by a bicarbonate anion, which originates from the dissolved CO_2 , a hydroxide anion or even self-compensation from the carboxylate groups, according to the following reaction.



This process relies on the oxidation of the P(2CAT-K) polymer by O_2 to form H_2O_2 , but also simultaneously depletes the reductive properties of the conjugated polymer. However, the oxidation of P(2CAT-K) is a reversible process, since the electron transfer process involves only the formation of polarons or

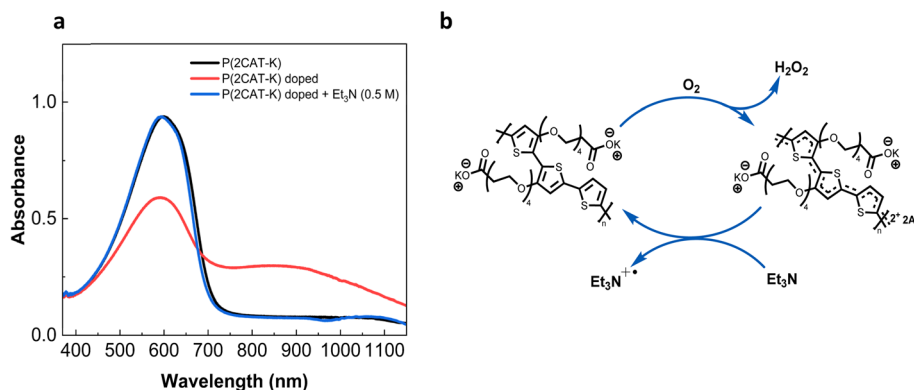


Fig. 3 (a) UV-Vis absorption spectra of a freshly prepared polymer aqueous nanodispersion (black line) sparged with O_2 (red line) and then mixed with 0.5 M of Et_3N (blue line). The disappearance of the polaron peak and the recovery of the $\pi-\pi^*$ transition peak shows the reversibility of the polymer oxidation by addition of the Et_3N reducing agent. (b) Scheme of the polymer doping/dedoping cycle. P(2CAT-K) reduces O_2 to H_2O_2 and then it is regenerated by oxidizing Et_3N to Et_3N^{+} .



bipolarons. This contrasts with the irreversible change in the chemical structure that is found in other molecules that are also able to reduce O_2 to H_2O_2 , such as ascorbic acid and phenols. To prove the reversibility of this process, triethylamine (Et_3N), a known reducing agent, was used to test the reversibility of the P(2CAT-K) oxidation by O_2 . As shown in Fig. 3, a fresh polymer solution (black line) was sparged with O_2 for 30 min, which resulted in doping of the P(2CAT-K) (red line), as evidenced by the formation of the polaron peak in the near-infrared region. Then, the addition of Et_3N led to the regeneration of the neutral polymer: the polaron peak disappeared and the $\pi-\pi^*$ transition peak was completely recovered. Moreover, the same results were obtained repeating the same doping and dedoping procedure with the same polymer solution with added Et_3N , except for a lower obtained doping level (see Fig. S23). Further experiments are needed to optimize this process, but these preliminary results already highlight the possibility of repetitive doping/dedoping redox cycles and thereby continuous H_2O_2 production.

Conclusion

A novel carboxylate-functionalized polar polythiophene, P(2CAT-K), was synthesized. P(2CAT-K) displays a low IE^0 of -4.4 eV that drives the ORR of O_2 to H_2O_2 in water without the need for an external electron source and low pH. The considerable amount of H_2O_2 produced by P(2CAT-K) as well as the reversible nature of the reaction could allow for new applications that are driven by autonomous H_2O_2 production with CP, such as antiseptic coating materials. We believe that the high level of H_2O_2 that is produced spontaneously *via* ORR in water by P(2CAT-K) could indicate that other low IE CPs will produce H_2O_2 under similar conditions.

Author contributions

R. K. and C. B. conceived the study and wrote together with help from all co-authors. A. Y. S., with input from J. H., carried out the synthesis and structural characterization of reaction intermediates, monomers and polymers. C. B. performed the UV-Vis spectroscopy, cyclic voltammetry and qualitative and quantitative analysis of H_2O_2 . Q. Z. performed UPS characterization. M. V. supervised the electrochemistry analysis, M. F. supervised the UPS characterization, and R. K. supervised the study.

Conflicts of interest

The authors declare no conflicts of interest.

Data availability

The data supporting this article has been included as part of the supplementary information (SI). Supplementary information is available. See DOI: <https://doi.org/10.1039/d5ta07162c>.

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