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# On the Use of Polyelectrolytes and Polymediators in Organic Electrosynthesis

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**Abstract:** Although organic electrosynthesis is generally considered to be a green method, the necessity for excess amounts of supporting electrolyte constitutes a severe drawback. Furthermore, the employment of redox mediators represents an additional separation problem. In this context, we have explored the applicability of soluble polyelectrolytes and polymediators using the TEMPO-mediated transformation of alcohols to carbonyl compounds as test reaction. Catalyst benchmarking based on CV studies indicates that the redox-active polymer can compete with molecularly defined TEMPO species. While a high efficiency of the alcohol oxidation was achieved in preparative-scale electrolysis, our polymer-based approach allows for the separation of both mediator and supporting electrolyte in a single membrane filtration step. Moreover, we have shown that both components can be reused for multiple cycles.

Compared to classical methods for the oxidation and reduction of organic compounds, organic electrosynthesis offers several economic, ecologic and practical advantages. The use of electric current instead of redox reagents provides the opportunity for reducing the overall cost, waste generation and energy consumption of a process.<sup>[1]</sup> However, despite the substantial progress that has been made in recent years, the potential of the method has yet to be fully exploited and expressed. This is in part due to the fact that the following issue has been neglected for a long time: Although organic electrosynthesis is generally considered as a green method, the employment of excess amounts of supporting electrolyte constitutes a severe drawback. Thus, following the completion of a reaction, the supporting electrolyte must be separated from the reaction mixture. Unless it is recovered and reused, it constitutes a source of waste. More specifically, tetraalkylammonium salts are often employed as supporting electrolytes, since they provide good solubility in organic solvents and high electrochemical stability.<sup>[2]</sup> However, the good solubility often leads to difficulties with separation of the salts, and in many cases column chromatography is required for this purpose. Since the supporting electrolyte represents a source of waste and a key-expense factor, the reuse of the separated salts for multiple cycles is highly desirable for industrial applications.<sup>[3]</sup> A further separation issue is caused by redox mediators, which are often used to facilitate electron transfer and to avoid electrode passivation.<sup>[4]</sup> The simplification of the separation of both supporting electrolyte and mediator from the reaction medium would certainly make electroorganic synthesis a more appealing synthetic tool.

The separation issue was addressed by Fuchigami et al., who

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successfully applied silica gel supported piperidinium salts as dispersed-phase supporting electrolytes for a number of electrochemical transformations.<sup>[5]</sup> A more recent approach by Little et al. comprises a polymeric ionic liquid/carbon black composite, which is recovered after completed electrolysis by centrifugation.<sup>[6]</sup> Attempts for the immobilization of mediators on microparticles, such as poly(styrene)-supported phenyl iodide<sup>[7]</sup> and TEMPO on silica gel,<sup>[8,9]</sup> have also been reported. While all these dispersed-phase approaches facilitate the separation and reuse of the supporting electrolyte, they are associated with drawbacks such as low ionic conductivity and poor kinetics of the electron transfer between electrode and mediator unit. In fact, halide salts are needed as co-mediators in order to allow for activation of the solid-supported mediator units.<sup>[7–9]</sup>

Compared to the dispersed-phase strategy, the attachment of mediator and/or supporting electrolyte to soluble polymer backbones (polymediators and polyelectrolytes) to create a homogeneous electrolyte represents a promising approach for improving both ionic conductivity and electron transfer kinetics. To date, this has only been attempted by Steckhan et al., who tested a polymer-supported triarylamine for the oxidation of anisyl alcohol.<sup>[10]</sup> While in principle, activity of the polymediator was observed, the system still suffered from poor electron transfer kinetics and complete degradation of the soluble polymer in the course of electrolysis. This study prompted us to investigate the requirements for a successful use of soluble polymers in electrosynthesis, and to develop a more robust and active system, which allows for practical current densities and repeated use. Herein we report the first protocol for the selective electrooxidation of alcohols to carbonyl compounds using recyclable polyelectrolytes and polymediators in combination with membrane separation processes for recovery of the polymers (see Scheme 1).



**Scheme 1.** Concept for the use of soluble polymediators and polyelectrolytes in electrosynthesis.

With respect to the redox-active unit, we selected 2,2,6,6-tetramethylpiperidin-1-oxyl (TEMPO), a robust, versatile and well-characterized mediator for homogeneously catalyzed electrochemical transformations.<sup>[11,12]</sup> To construct suitable

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homo- and copolymers for our application, we have modified a procedure from Schubert et al. for the synthesis of redox-active polymers applied in redox-flow batteries.<sup>[13]</sup> The resulting structures **HP-1** and **HP-2** (Figure 1) contain redox-active TEMPO moieties and electrochemically stable alkylammonium perchlorate units covalently attached to a poly(methacrylic acid) backbone. Each polymer is accessible from commercially available starting materials in only two straightforward and scalable steps (see SI).



Figure 1. Structures of polyelectrolyte HP-1 and polymediator HP-2.

For optimized solubility and diffusion properties of **HP-2**, we adjusted  $M_w$  to about 2700 g mol<sup>-1</sup> (corresponds to ~11 monomer units) by using a chain transfer additive in the radical polymerization of the methacrylic acid derivatives. Initially, we also studied copolymers with different ratios between redoxactive and supporting electrolyte units. Since these polyelectrolyte mediators are significantly less redox-active compared to **HP-2** (for more details see SI), we focussed our efforts on the homopolymer-based approach.

For the electrochemical characterization of HP-2, cyclic voltammetry was carried out in a 0.1 M solution of NBu<sub>4</sub>ClO<sub>4</sub> in CH<sub>3</sub>CH/H<sub>2</sub>O (95:5), whereby the polymer content was adjusted to a 1 mM concentration of the redox-active repeat unit. The CV of HP-2 (blue line in Figure 2, left) shows reversible redox behavior at  $E_0 = 0.44$  with a 140 mV anodic shift compared to free TEMPO (black line), which we used as a benchmark for the electroanalytical studies. In each case, plots of the peak current density  $i_{\rm P}$  vs. the square root of the scan rate v show a linear dependency (see Figure 2, top right), indicating that both for TEMPO and HP-2 the charge transfer is similar to a process controlled by linear diffusion.<sup>[14]</sup> The diffusion coefficient D<sub>HP-2</sub> was calculated from the slope of  $j_P$  vs.  $v^{0.5}$  according to the Randles-Sevcik treatment (for details see SI). With 8.8-10<sup>-6</sup> cm<sup>2</sup>  $s^{-1}$ , this value is somewhat lower as compared to  $D_{TEMPO}$  with 3.2 10<sup>-5</sup> cm<sup>2</sup> s<sup>-1</sup>, whereby the latter value is in good agreement with the literature.<sup>[15]</sup> The decrease in D follows the behavior predicted by the Stokes-Einstein equation in the sense that diffusion is curbed with increasing molecular weight. The ratio between  $D_{HP-2}$  and  $D_{TEMPO}$  suggests that the redox-centers are non- or weakly interacting and that the transport mechanism during the electrochemical conversion at the electrode is best described as classic diffusion (for more details see SI).[16]

Upon increasing the concentration of redox active repeat units, a linear increase of the peak current density  $j_P$  is observed (see Figure 2, bottom right), which confirms the diffusion-type limitation of the electron transfer. Noteworthy, the separation between anodic and cathodic peak potential ( $\Delta E_P$ ) is significantly smaller for **HP-2** as compared to TEMPO. While  $\Delta E_P$  for TEMPO

is with 70 mV relatively close to the 57 mV expected for a reversible single electron transfer in a diffusion controlled process (whereby both  $E_0$  and  $\Delta E_P$  are in good agreement with the literature),<sup>[17]</sup>  $\Delta E_P$  is only 35 mV for **HP-2**. This is a significant deviation from the behavior of other linear polymers in solution having ferrocene or anthraquinone units attached to the chain, where  $\Delta E_P$  values of 60 mV and higher were reported for the same scan rate.<sup>[16,18]</sup> While further investigation of this unusual behavior is ongoing, we can conclude from the results presented in Figure 2 that **HP-2** renders diffusion-controlled current densities which are *i*) of the same order as for TEMPO and therefore promising for application in indirect electrosynthesis and *ii*) adjustable by variation of the concentration of the redoxactive repeat units.



**Figure 2.** Left: Cyclic voltammetry of TEMPO and **HP-2** ([TEMPO unit] = 1 mM, electrolyte: 0.1 M NBu<sub>4</sub>ClO<sub>4</sub> in CH<sub>3</sub>CN + 5 vol% H<sub>2</sub>O, WE = glassy carbon, RE = Ag/0.01 M AgNO<sub>3</sub>), v = 50 mV s<sup>-1</sup>. The current density *j* has been normalized with respect to the peak current density *j*<sub>P</sub>. Right: Plot of the peak current densities *j*<sub>P</sub> (with background correction) vs.  $v^{0.5}$  and *c*, respectively.

 Table 1. Comparison between TEMPO and HP-2 with regard to redox behavior and mass transport.

Mediator	<i>E</i> ₀ [V] <sup>[a]</sup>	<i>∆E</i> <sub>p</sub> [mV] <sup>[a]</sup>	D [cm <sup>2</sup> s <sup>-1</sup> ] <sup>[b]</sup>
TEMPO	0.30	70	3.2·10 <sup>-5</sup>
HP-2	0.44	35	8.8·10 <sup>-6</sup>

[a] Determined at v = 50 mVs<sup>-1</sup>, [TEMPO unit] = 1 mM. [b] Determined via Randles-Sevcik treatment (see Figure 2, top right and SI).

We continued our investigation with characterizing the electrocatalytic behavior of **HP-2**. The voltammetric profiles recorded at 50 mVs<sup>-1</sup> in absence and presence of benzylalcohol and *N*-methyl imidazole (NMI, employed as proton scavenger) are depicted in Figure 3 (left). In presence of alcohol and NMI, **HP-2** shows a pronounced catalytic wave without cathodic peak in the reverse scan. Interestingly, the ratio between the maximum current densities with and without substrate ( $j_{cat}/j_{P}$ , reflecting the rate of the homogeneous reaction), is significantly higher compared to molecular TEMPO under the same conditions. A plot of  $j_{cat}/j_{P}$  vs. [PhCH<sub>2</sub>OH] is shown in Figure 3 (bottom right) for both **HP-2** and TEMPO at a 1 mM concentration of mediator units, whereby a linear dependency is

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observed in each case. These plots show that *i*) the reaction is first order with regard to the alcohol substrate,<sup>[19]</sup> and that *ii*) **HP-2** exhibits the superior homogeneous rates in the studied concentration regime.



**Figure 3.** Left: Cyclic voltammetry of **HP-2** ([TEMPO unit] = 1 mM) in absence and presence of 0.1 M PhCH<sub>2</sub>OH and 0.45 M NMI (experimental conditions: see Figure 2). Top right: Plot of the maximum catalytic current  $j_{cat}$  vs. the square root of the concentration of PhCH<sub>2</sub>OH (v = 50 mVs<sup>-1</sup>). Bottom right: Plot of  $j_{cat}$  vs. v (0.1 M PhCH<sub>2</sub>OH and 0.45 M NMI).

For a quantitative treatment of the kinetics of the alcohol oxidation, we determined the homogeneous rate constants  $k_{cat}$ (see Table 2) using CV under purely kinetic control and pseudo first-order conditions (see Figure 3, bottom right).<sup>[19]</sup> Both for TEMPO and for HP-2, jcat reaches a plateau value (jmax) at sufficiently high v. This behavior allows for the extraction of  $k_{cat}$ , which is about 60 times higher for HP-2 compared to TEMPO (for details see SI). The improved rate of the homogeneous reaction in the case of HP-2 can be explained by the electron withdrawing substituent in position 4 of the TEMPO unit (i. e. the acyloxy linker) and the resulting anodic shift of  $E_{0x}$ . Similar behavior was recently reported by Stahl et al., who found that the electrocatalytic activity of N-oxyl radicals rather depends on the mediator potential than on steric influences.[11] It is important to highlight that the results presented in Table 2 only reflect the reaction rate under kinetic control, and that a scenario where mediator transport is rate-limiting (for instance at low alcohol concentrations) would turn the outcome in favor of moleculardefined TEMPO due to its higher diffusion coefficient.

Table 2. Comparison of the electrocatalytic behavior of HP-2 and TEMPO.

Mediator	j <sub>max</sub> [mA cm <sup>-2</sup> ] <sup>[a]</sup>	<b>k</b> cat <b>[S</b> <sup>-1</sup> ] <sup>[b]</sup>	
TEMPO	1.8	5	
HP-2	7.0	301	

[a] Catalytic current for the oxidation of PhCH<sub>2</sub>OH determined under pseudo first-order conditions (see Figure 3, bottom right). [b] Pseudo-first order rate constant for the oxidation of PhCH<sub>2</sub>OH, calculated from *j*<sub>cat</sub> (for details see SI).

While the results presented above clearly indicate that dissolved polymediators allow for efficient electrocatalysis, it is also important to consider the properties of the polyelectrolyte **HP-1**. Since the electrolyte resistance renders a major contribution to

the cell voltage in a bulk electrolysis, a sufficiently high ionic conductivity  $\sigma$  is crucial. We have therefore determined  $\sigma$  in CH<sub>3</sub>CN at different concentrations (Figure S22). A comparison to NBu<sub>4</sub>ClO<sub>4</sub>, a typical benchmark salt, shows significantly decreased values for **HP-1** (for instance 2.5 mS cm<sup>-1</sup> for a solution containing 0.3 M repeat units as compared to 9.6 mS cm<sup>-1</sup> for a solution of NBu<sub>4</sub>ClO<sub>4</sub> of the same concentration of supporting electrolyte units). However, the conductivity of **HP-1** is still in the appropriate range for bulk electrolysis.

With the analytical data in hand, we elaborated a protocol for preparative scale electrolysis. The optimized conditions comprise an electrolyte consisting of HP-1 (0.05 M supporting electrolyte units), 1.5 - 5 mmol alcohol substrate, HP-2 (15 mol% mediator units) and 2 mmol lutidine dissolved in CH<sub>3</sub>CN. In order to facilitate H<sub>2</sub> evolution as cathodic half-reaction, we added H<sub>2</sub>O (5 vol%) and used a Pt cathode. A comparison between different anode materials revealed that inexpensive carbon roving gives the best results. Bundled into a brush-type electrode (see Figure S20), it provides a high surface area while passivation during electrolysis is minimized compared to platinum, glassy carbon and graphite. An inexpensive size-exclusive membrane was used as a separator to prevent discharge of the polymediator at the cathode. Using this set-up, the conversion of several benzylic, allylic and aliphatic alcohols was carried out at 0.56 V vs. Ag/AgNO<sub>3</sub> (see Table 3).

Table 3. Scope of the electrocatalytic alcohol oxidation using HP-1 as supporting electrolyte and HP-2 as mediator.  $^{\rm [a]}$ 

Product <sup>[b]</sup>	Controlled E <sup>[c]</sup>			Controlled <i>i</i>		
	Q [F]	<b>FE [%]</b> <sup>[d]</sup>	Q [F]	<i>i</i> [mA]	<b>FE</b> [%] <sup>[d]</sup>	
	1.8 2	94 87 (84) <sup>[e]</sup>				
O <sub>2</sub> N O	1.8	84	1.8 1.9 2.0	4.5 4.5 4.5	92 (49) <sup>[f]</sup> 87 (83) <sup>[e]</sup> 83	
<i>t</i> Bu O	1.8	89				
0	1.8	97				
€ N S S S S S S S S S S S S S S S S S S	1.8	58				
	1.8	84				
	1.8	94	2 1.9	4.5 6.5	100 (3) <sup>[f]</sup> 79 <sup>[g]</sup>	
	1.8	89	1.8	3.0	94 (8) <sup>[f]</sup>	
~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	1.8	65				
	1.8	96				
	1.8	88	1.8	3.0	87 (n. d.) <sup>[f]</sup>	

0 3.6 64<sup>[b]</sup>

[a] Conditions: r. t., divided cell (separator: regenerated cellulose with 1 kDa MW cut-off), anode: carbon roving, cathode: Pt sheet, solvent: CH<sub>3</sub>CN/H<sub>2</sub>O (95:5 vol/vol). Anolyte: 20 mL solvent containing 54 mg HP-2 (15 mol% mediator units), 313 mg HP-1 (corresponds to 0.05 M supporting electrolyte units), 250  $\mu$ L 2,6-lutidine, 1.5 mmol substrate. Catholyte: 20 mL solvent containing 313 mg HP-1. [b] The starting material is the corresponding alcohol (pentane-1,5-diol for δ-lactone). [c] E = 0.56 V vs. Ag/AgNO3. [d] Faradaic efficiencies are based on the yield determined with GC (internal standard). [e] Isolated yield. [f] Electrolysis in absence of HP-2. [g] 5 mmol batch size.

The Faradaic efficiencies (FE) were calculated on the basis of the yields determined via GC (internal standard) after passing 1.8 - 2 F charge per mole of substrate, and are located between 64% and 97%. In addition, product isolations were carried out in the case of *p*-methoxy benzaldehyde and *p*-nitro benzaldehyde, whereby comparable FEs were obtained. In order to demonstrate the scalability, we have converted 1.5 g 1-octanol using the same cell and electrolyte volume, whereby 1-octanone was obtained in 75% FE (for experimental details see SI). We have also tested some of the reactions under controlled current conditions (see Table 3, right), and found that the FEs are similarly useful (72% - 100%). To confirm the active role of the mediator under these conditions, control reactions in absence of HP-2 were carried out, which rendered significantly lower FEs (0 - 48%, see footnote f). Moreover, we have tested the selectivity for conversion of primary vs. secondary alcohols in a competition experiment: In presence of an equimolar amount of 2-octanol, 1-octanol is converted with >99% selectivity (see SI). Recycling studies indicate that HP-1 and HP-2 can be used for at least 5 runs without loss of activity and conductivity, when dialysis is applied as separation method between each run (see Figure 4, green bars). Recovery of the polymers via ultrafiltration leads to a stable performance for three runs (Figure 4, red bars), whereby a loss of activity is observed in the fourth run (probably due to a non-quantitative retention in the filtration process).



Figure 4. Recycling study for the use of HP-1 and HP-2 in the synthesis of octane-1-one (galvanostatic electrolysis, conditions: see Table 3).

In conclusion, we have demonstrated the feasibility of using soluble polyelectrolytes and polymediators in electrosynthesis for an efficient recovery of both components by membrane separation processes. In the particular case of polymer-bound TEMPO (HP-2), we found that compared to free TEMPO the rate of the alcohol oxidation is significantly improved. The superior homogeneous kinetics can in part compensate the unfavorable diffusion coefficient of HP-2, ultimately leading to competitive current densities in the electrolysis. We expect that our findings will have an important impact on future developments of polyelectrolyte-polymediator systems, and are

therefore currently expanding our concept to other electrochemical transformations and mediator units.

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**Keywords:** Electrosynthesis • TEMPO • Polyelectrolyte • Redox Mediator • Ultrafiltration

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#### Entry for the Table of Contents (Please choose one layout)

Layout 1:

## COMMUNICATION

**Combining electrosynthesis with membrane filtration.** The attachment of TEMPO mediator and supporting electrolyte to soluble polymers enables a selective anodic conversion of alcohols to carbonyls and allows for efficient recovery/recycling of the mediator and the salt by membrane filtration.



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Page No. – Page No.

On the Use of Polyelectrolytes and Polymediators in Organic Electrosynthesis