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An Iodide-Mediated Anodic Amide Coupling

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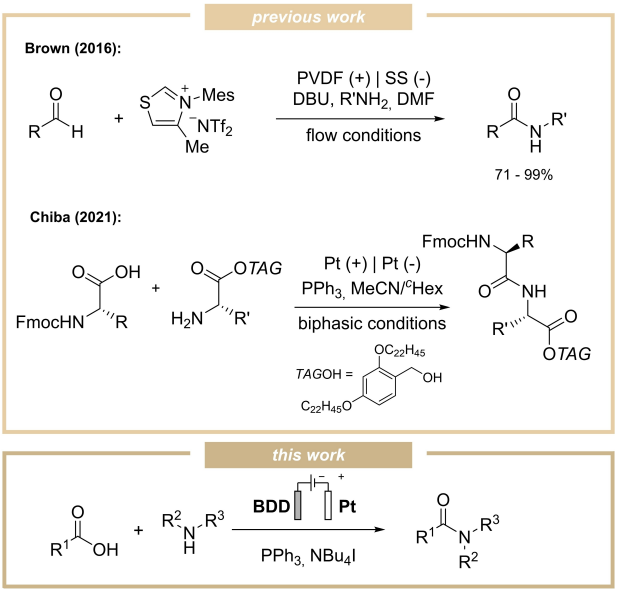
Abstract: The ubiquity of amide bonds, present in natural products and common pharmaceuticals renders this functional group one of the most prevalent in organic chemistry. Despite its importance and a wide variety of existing methods for its formation, the latter still can be a challenge for classical activating reagents such as chloridating agents or carbodiimides. As the spent reagents often cannot be recycled, the development of more sustainable methods is highly desirable. Herein, we report an operationally simple and mild indirect electrochemical protocol to effect the condensation of carboxylic acids with amines, forming a wide variety of carboxamides.

Introduction

The amide bond is amongst the most prevalent structural motifs found in natural products, it represents the key linkage in proteins and is encountered in high value products like polymers or pharmaceuticals.[1–2] Although the importance of this moiety is well recognized, amide bond formation remains a contemporary challenge in organic chemistry as highlighted by the ACS Green Chemistry Institute Roundtable (ACS GCIPR), who called out for "amide bond formation avoiding poor atom economy".[3] Well established methods to forge amide bonds typically rely on pre-activation of carboxylic acids by converting them into a more reactive intermediate like an active ester, mixed anhydride or acid chlorides. Additionally, the utilization of coupling reagents like carbodiimides or uronium salts is a common and well-explored methodology to form amides from carboxylic acids and amines.[2,4] However, indirect amide bond formations of this kind are cumbersome, expensive, and waste-intensive, hence more ecological and economical methodologies are desirable. Alternative approaches starting from carboxylic acid surrogates like alkynes,[5–9] alcohols[10–12] or aldehydes[13–16] have been reported, which all utilize a redox approach to form the amide group. Among these approaches, the Brown group recently presented an electrochemical, microfluidic system using a N-heterocyclic carbene to transform aldehydes into the respective amide via a Breslow intermediate and subsequent oxidation (see Scheme 1).[16] A recent growing interest in electrochemical transformations has led to the use of electric current as a pseudo-reagent in such reactions, however, electrochemical methods for direct condensation of a carboxylic acid and amine remain scarce.[17–22] In 1991, the group of Ohmori reported a procedure, using anodically oxidized PPh_3 to activate carboxylic acids for ester and amide synthesis. However, this methodology is limited to carboxylic acids and primary, aliphatic amines with oxidation potentials more positive than that of PPh_3. Additionally, the selected electrolyte system, containing DCM and lutidinium perchlorate, raises safety and environmental concerns and therefore might not be favorable today.[18]



Scheme 1. Synthesis of amides utilizing electric current as clean oxidant.

The group of Chiba published a biphasic electrochemical peptide synthesis which also utilizes PPh_3 as suitable coupling reagent for direct condensation of an *N*- and a *C*-protected amino acid (see Scheme 1). Although *C*-protected α-amino acids are usually oxidized at higher potentials relative to PPh_3, the authors introduced a highly lipophilic benzylic alcohol as protecting group for the *C*-terminus to enhance its solubility in non-polar solvents like cyclohexane. This allowed the electro-

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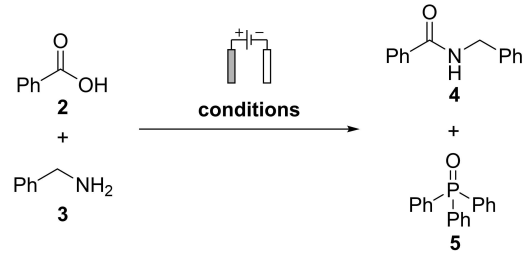
chemical oxidation to be performed in a biphasic system, which not only circumvented the risk of unselective oxidation but also facilitated the recovery of the product and Ph_3PO from the reaction mixture.[21] However, this soluble tag-assisted electrochemical method was only demonstrated for amino acids and requires pre-functionalization of the substrates. Thus, easily oxidizable amines remain challenging substrates for an anodic amide bond formation. While the above-mentioned methods produce Ph_3PO as a stoichiometric co-product, its reduction back to PPh_3 has been achieved by various methods, including an electrochemical one developed by Tanaka and Sevov, which is also applicable on a large scale.[23–33] Unlike co-products formed in amide bond formations with traditional coupling reagents, Ph_3PO has proven to be an easily recyclable spent coupling reagent, and thus provides potential access to a greener amide coupling.

Herein, we report an electrochemical condensation method inspired by the Garegg-Samuelsson reaction and the work of Leopold Horner to form amides from carboxylic acids and amines.[34–35] We envisioned a related electrochemical approach, eliminating the need for stoichiometric oxidants to activate the triphenylphosphine, while maintaining high tolerance to challenging substrates.

Table 1. Variation of the optimized reaction conditions.



Entry	Conditions	Yield [%][a]
1	**Starting conditions**: $C_{gr}(+)\|Pt(-)$, BzOH (1.0 equiv.) $BnNH_2$ (1.1 equiv.), Bu_4NI (1.1 equiv.), PPh_3 (1.1 equiv.), MeCN (0.05 M), $j = 8$ mA cm^{-2}, $Q = 2.2$ F	56
2	$J = 12$ mA cm^{-2}	81
3	Glassy carbon instead of graphite	81
4	BDD instead of graphite	84
5	NEt_4I instead of NBu_4I	71
6	NaI instead of NBu_4I	59
7	10 mol% NBu_4I	54
8	**1.5 equiv. PPh_3 instead of 1.1 equiv.**	**98 (88)**
9	No current	0
10	No PPh_3	0
11	No iodide	18

[a] Determined by ^{1}H NMR with 1,4-bis-TMS-benzene as internal standard, isolated yield in parentheses.

Results and Discussion

Initially, we used a model system in an attempted direct activation of triphenylphosphine (**1**) by anodic oxidation. Although a direct anodic oxidation is indeed feasible, the relatively high oxidation potential of 1.00[36]–1.06[37] V vs. SCE limits the scope of appropriate amines to be employed in this method. In order to realize a one-pot procedure which does not affect either of the two coupling partners, iodide was introduced as a redox mediator, since molecular iodine is known to be a suitable activating agent for PPh_3.[34]

Therefore, the use of an iodide containing supporting electrolyte proved to be beneficial, in order to lower the required electrode potential, since the oxidation of iodide proceeds smoothly at +0.26 V vs. SCE.[38]

The feasibility of this new approach was investigated using benzoic acid (**2**, 1.0 equiv.) and benzylamine (**3**, 1.1 equiv.) in the presence of NBu_4I (1.1 equiv.) and PPh_3 (1.1 equiv.). To carefully exclude any form of oxygen, the reaction was run under an argon atmosphere in dry MeCN (0.05 M). This mixture was electrolyzed using a combination of a graphite anode and a platinum cathode passing 2.2 F per mole **2** at a current density of $j = 8$ mA cm^{-2} giving the desired product **4** in 56% yield (see Table 1) together with the expected co-product triphenylphosphine oxide (**5**, TPPO).

Screening different current densities (see Table S1) showed, that an increase did not affect the yield up to $j = 11$–12 mA cm^{-2} which gave a significantly higher yield of 77% and 81% respectively. Increasing the current density even further resulted in decreasing yields. Therefore, a current density of 12 mA cm^{-2} (at 2.1 cm^2 immersed electrode surface) was identified as the optimum value.

While the use of standard graphite electrodes already gave a satisfactory yield of 81%, different anode materials were also examined for their performance in this reaction (see Table S2). Among the metal-based materials tested, platinum performed best with 75% yield, but could not exceed the efficiency of the graphite electrode. Testing different carbon variants revealed glassy carbon to perform equally well and therefore should be preferable due to its higher chemical inertness.[39] However, the high performance material BDD (boron doped diamond) showed the best result of all tested anode materials, providing the desired product in 84% yield. Based on the highest yield, together with the outstanding electrochemical properties of BDD electrodes (widest solvent window, low electrode fouling, stability towards extreme conditions),[40] further work was continued with BDD as the optimal anode material. However, it should be noted that a more economic setup can be based on the cost-efficient graphite anode.

During the optimization process, different organic and inorganic iodide salts were examined as supporting electrolytes (see Table S3). For the evaluation of the optimal salts, not only on the overall yield but also the process mass intensity (PMI) of the reaction was taken into consideration. While promising, yet uncommon supporting electrolytes like the room temperature ionic liquid (RTIL) 1-butyl-3-methylimidazolium iodide [BMIM]I, neither gave high yields nor desirably low PMIs, the initially chosen NBu_4I proved to give the highest yield whilst at the same time providing the lowest PMI value of 94. It is noteworthy that the inexpensive inorganic iodide salt NaI still gave 59% yield, despite its relatively low solubility. The application of an RTIL as both, supporting electrolyte and (co-)solvent would have been beneficial, since purification and recycling of the iodide source is easy to achieve by a simple washing and drying procedure as

presented by Bornemann and Handy[41] Nevertheless, NBu_4I could also be recoverd by crystallization from EtOAc.

A critical parameter in classic amide bond formations is the choice of the appropriate solvent. According to Sheppard et al., DMF and DCM are the most prevalent solvents for amide coupling on both laboratory and industrial scale, despite severe health and environmental concerns attributed to them.[42] We therefore aimed to identify a suitable reaction solvent in line with common solvent selection guides like that of Prat et al. (see Table S4).[43] Unfortunately, all tested "green" solvents like acetone, ethyl acetate, *tert*-butanol, methanol or dimethyl carbonate underperformed and resulted in a maximum of 3% of the desired product, which could attributed mainly to low conductivity of the resulting solution. In comparison, less eco-friendly solvents like THF, DMF and DCM were tested as well. These solvents gave significant amounts of product, but the yields still could not compete with the performance of acetonitrile. Therefore, we identified the latter as the optimal solvent, improving the "greenness" even further, since acetonitrile can be derived from ethanol and ammonia through ethylamine as an intermediate.[44–46]

Further optimization studies regarding the utilized phosphine, the overall concentration, the supporting electrolyte loading and the applied charge did not result in any changes of the optimized reaction conditions. However, decreasing the iodide loading to 10 mol% still gave the desired product in 54% yield. The decreased yield might be attributed to a mismatch of the high current density and the low concentration of iodide, meaning the electrons may be transferred at a faster rate than the iodide being regenerated.

The last crucial parameter to be tested was the loading of PPh_3 (see Table S9). We observed that a slight increase of the PPh_3 loading from 1.1 equiv. to 1.5 equiv. resulted in a significant increase of the coupling yield to 98%. To account for the larger amount of phosphine, we simultaneously adapted the applied charge to 3.3 F (equals 2.2 F/mol relative to PPh_3). To verify this result, we applied the optimized parameters to a 1 mmol scale electrolysis, which, after column chromatography, furnished the desired product in 88% isolated yield (98% NMR yield).

With the optimized reaction conditions in hand, the substrate scope of this reaction was investigated (see Scheme 2).Initially, it should be proven that this methodology is not limited to aromatic carboxylic acids like benzoic acid, but is also applicable to aliphatic acids. Therefore, phenylacetic acid was chosen as a particularly challenging substrate since it is not only aliphatic but also prone to oxidative decarboxylation resulting in the formation of a stable benzylic radical. Gratifyingly, the amide coupling with benzylamine proceeded smoothly to give the desired product **8** in 71% isolated yield without any side product detected, that resulted from decarboxylative radical formation. Even the smallest aliphatic acids formic, acetic, propionic and trifluoroacetic acid gave their respective anilides **28**–**31** in 72–92% yield. It was also found that amide **34** was accessible in a high yield by applying the electrochemical procedure, while conventional coupling reagents such as HATU failed to give **34** in a satisfactory yield.

Additional investigation of the acid scope also proved *N*-protected amino acids to be suitable acid components for this reaction, as were different heteroaromatic carboxylic acids and the pharmaceutical ibuprofen. Halogenated benzoic acid derivatives also gave the respective amides **6** and **7** in moderate yield, however, trace amounts of dehalogenated products could be observed by LCMS analysis.

Interestingly, 4-methoxycinnamic acid gave 54% of a mixture of the saturated and unsaturated amide in a 3:1 (sat.:unsat.) ratio. We initially ascribed this observation to the hydrogen evolution at the platinum cathode and therefore changed the cathode to the high overpotential material BDD ($\eta_{HER} = -1.1$ V)[47] to circumvent this problem. It turned out that the hydrogen evolution was not the problem, but rather a direct electron transfer from the cathode to the substrate, since the use of BDD exclusively produced the saturated amide **11** in 64% yield. The same phenomenon was observed with methacrylic acid which gave the respective amide **33** in a 1.7:1 ratio (sat.:unsat.). Non-conjugated alkene-moieties seem to be tolerated, as 6-heptenoic acid did yield the corresponding anilide **32** in 52% yield.

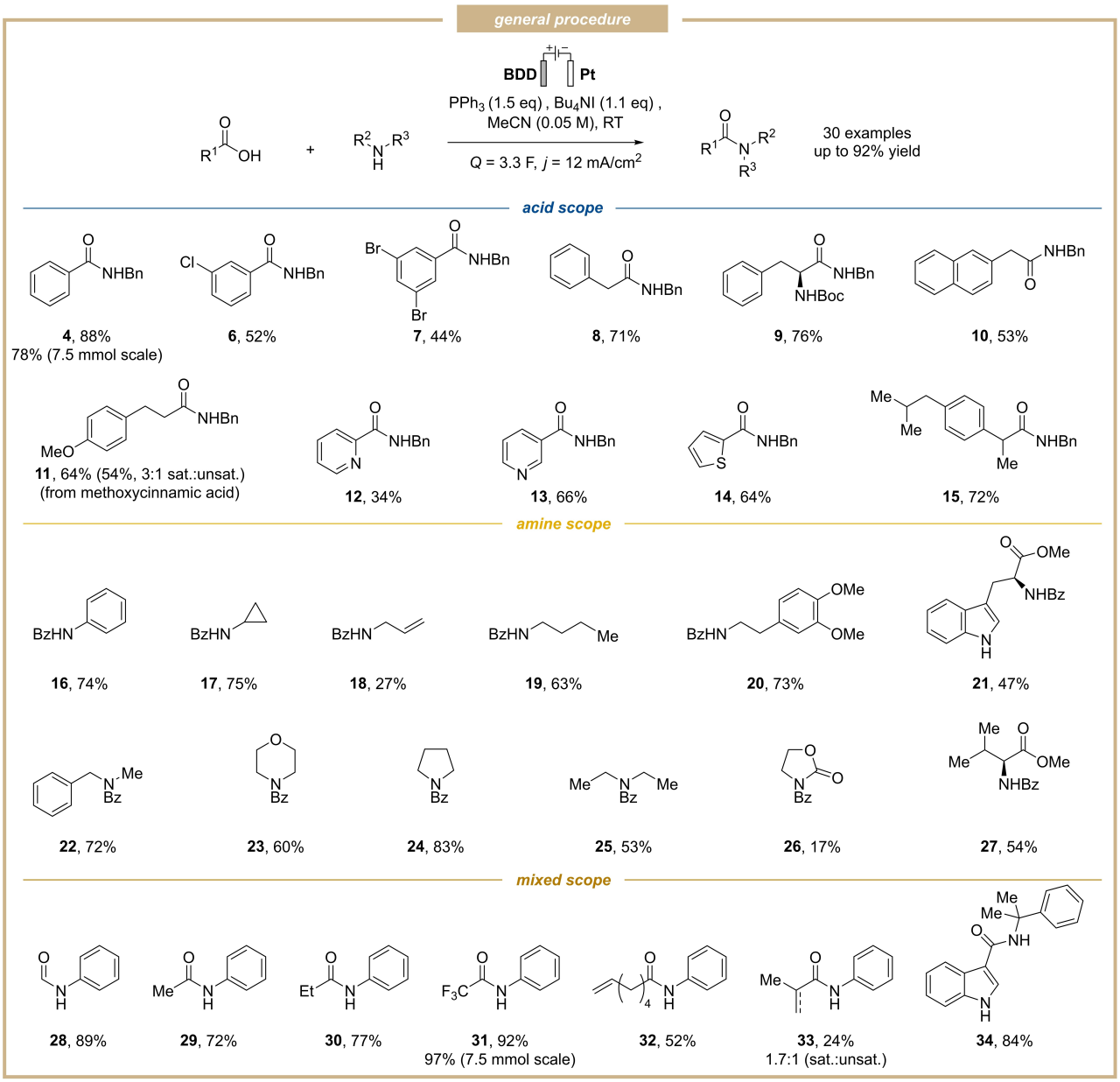
To test the scope of this method, a variety of different primary and secondary amines were investigated, which are typically prone to oxidation at low potentials. Aniline for example gave benzanilide **16** in a satisfactory yield of 74%, despite being known for forming polymers under anodic conditions.[48–49] Cyclopropylamine was also tolerated under the standard conditions to form amide **17** without any detectable ring opening. Allylamine however gave a rather low yield of the corresponding amide **18**, which might be due to cathodic decomposition of the protonated amine. In contrast, secondary amines performed even better than expected, giving moderate to high yields. Pyrrolidine for example gave its benzamide **24** in 83% isolated yield, despite having an oxidation potential as low as +0.89 V vs. SCE.[38]

A rather unusual coupling component, 2-oxazolidinone, was also subjected to the standard electrolysis conditions. Although yielding **26** in only 17%, the acylation of this poorly nucleophilic substrate is encouraging.

Additionally, α-amino acid esters proved to be suitable amine components, which led us to investigate the feasibility of peptide couplings (see Scheme 3). Since most *C*-protected amino acids are supplied as hydrochlorides and therefore may be partly insoluble in acetonitrile, we decided to modify the developed protocol by stirring all components with 1.1 equiv. of triethylamine for 15 min prior to addition of the *N*-protected amino acid and subsequent electrolysis.

With this slight modification, five dipeptides with different protecting groups could be synthesized. Common protecting groups in peptide chemistry are being tolerated. *N*-Protecting groups such as Boc, Cbz, Fmoc and *p*-Tos are stable under these conditions. Interestingly, the acid sensitive *tert*-butyl ester also gave a moderate coupling yield.

To further demonstrate the utility of the method, gram scale experiments with 7.50 mmol of acid have been conducted. The model substrate **4** was obtained in 78% yield after column chromatography, while 85% of the supporting electrolyte could be recoverd by simple crystallization from ethyl acetate prior to

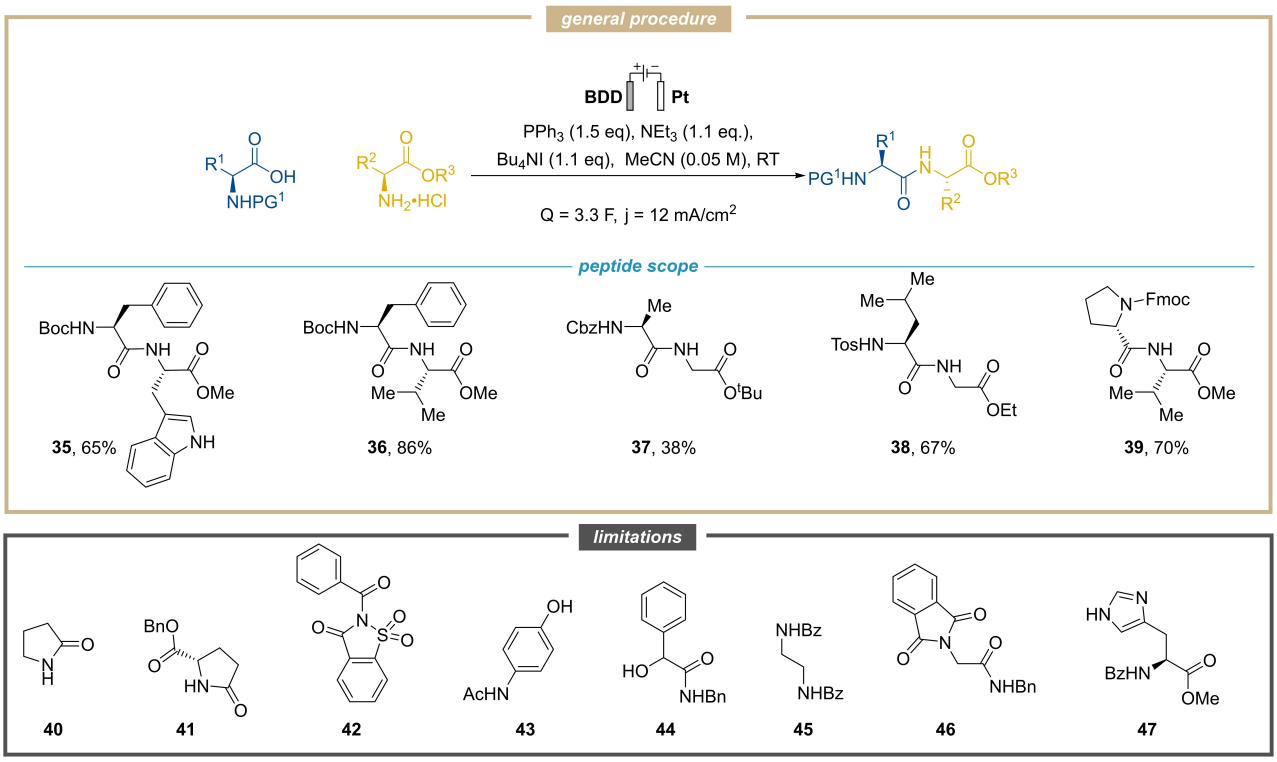


Scheme 2. Substrate scope for electrochemical amide bond formation. Reaction conditions: Acid (1.0 mmol), amine (1.1 mmol), PPh_3 (1.5 mmol), NBu_4I (1.1 mmol) in dry MeCN (20 mL), BDD-anode, Pt-cathode, Q = 3.3 F (318.4 C), j = 12 mA cm^{-2} (88.8 mA, 7.4 cm^2). All yields given are those of the isolated compound after chromatographic purification.[1] Yield using BDD instead of Pt as cathode material.

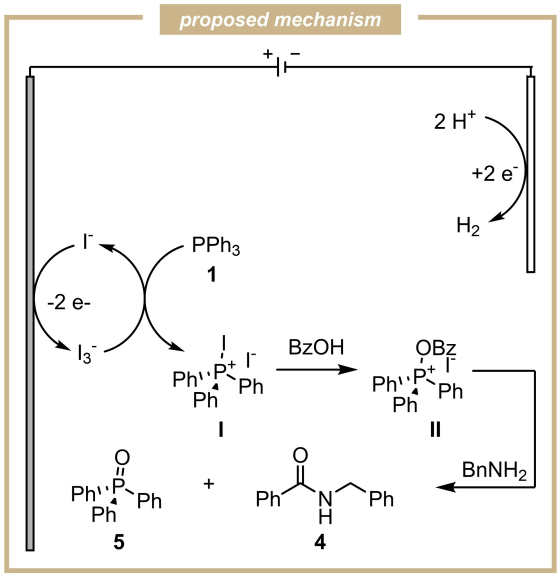
purification. Trifluoroacetanilide (**31**) was prepared on the same scale in 97% isolated yield.

Despite giving good results with the reported substrates, this procedure has shown some limitations, as depicted in Scheme 3. A detailed discussion of these limitations can be found in the Supporting Information.

Some studies were undertaken to investigate the reactions mechanism. Control experiments without current gave no product formation, thus proving that the reaction is driven by electrical current. This fact renders the procedure inherently safe, since no reactive reagents or elevated temperatures are need and the reaction can be halted immediately if necessary. Leaving out PPh_3 also resulted in no observed product, which proved its critical role in the mechanism. Changing the supporting electrolyte from NBu_4I to NBu_4PF_6 decreased the yield to 18% and simultaneously increased the cell voltage. This result validated that an iodide source is mandatory to keep the electrode potential low and thus the conditions mild. To identify the redox active species which interacts with PPh_3, cyclic voltammetry experiments were performed. We recorded the cyclic voltammogram of NBu_4I in MeCN (see Figure S2), which shows two reversible oxidation peaks at +0.35 V and +0.75 V vs. Ag/Ag^+. The first peak was ascribed to the oxidation of iodide to triiodide and the second to the subsequent oxidation of triiodide to



Scheme 3. Substrate scope for modified electrochemical amide bond formation and limitations. Reaction conditions: N-protected amino acid (1.0 mmol), C-protected amino acid (1.1 mmol), NEt_3 (1.1 mmol) PPh_3 (1.5 mmol), NBu_4I (1.1 mmol) in dry MeCN (20 mL), BDD-anode, Pt-cathode, Q = 3.3 F (318.4 C), j = 12 mA cm^{-2} (88.8 mA, 7.4 cm^2). All yields given are those of the isolated compound after chromatographic purification.



Scheme 4. Proposed mechanism for the electrochemical amide bond formation.

elemental iodine. To identify, which of the two iodine species interacts with PPh_3, cyclic voltammograms of this solution with increasing amounts of the phosphine were recorded, since an increase in current density from one of the anodic peaks was expected. Indeed, an increase of the first anodic peak with an increasing amount of PPh_3 could be observed, while the height of the second oxidation peak remained unchanged. This suggests triiodide to be the active iodine donor in this reaction to activate PPh_3. Based on these results, we propose the mechanism shown in Scheme 4 for the electrochemical amide bond formation using PPh_3. We assume that the initial step is the anodic oxidation of iodide to triiodide, which will undergo subsequent reaction with PPh_3 forming intermediate **I**. The latter will then undergo iodine displacement by one molecule of benzoic acid to form intermediate **II**, which in the next step undergoes aminolysis to form the desired amide **4** along with Ph_3PO as a potentially recyclable co-product.

Conclusion

An operationally simple and mild anodic protocol for indirect electrochemical amide bond formation has been developed. It utilizes commercially available PPh_3 as coupling reagent precursor and NBu_4I as redox mediator, to directly couple carboxylic acids with amines. The presented method was applicable to a broad substrate scope including oxidation labile amines, simple carboxylic acids and even α-amino acids tolerating common protecting groups from peptide chemistry.

Supporting Information

The Supporting Information includes detailed optimization studies, experimental procedures, mechanistic studies, and copies of NMR spectra.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords: amides · electrochemistry · iodine · peptide coupling · redox condensation

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