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RESEARCH ARTICLE OPEN ACCESS

Tuning the Efficiency of Electrocatalytic Primary Amine Synthesis via Reductive Amination of Levulinic Acid Using Pb as a Catalytic Additive

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Received: 29 October 2025 | **Revised:** 10 January 2026 | **Accepted:** 12 January 2026

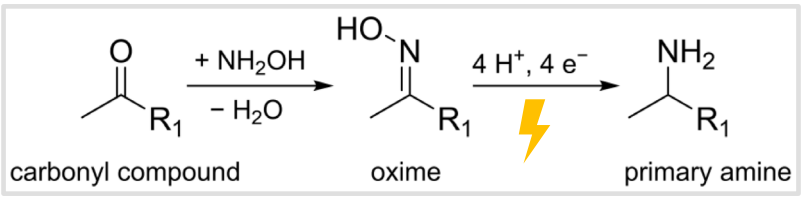
Keywords: amination | electrocatalysis | lead | oxime | reduction

ABSTRACT

Electrocatalytic reductive amination (ECRA) offers a sustainable approach for the synthesis of primary amines by replacing stoichiometric reducing agents with electrons and using water as both solvent and proton source. This atom-efficient strategy minimizes waste, avoids hazardous reagents, and eliminates the need for hydrogen handling. The coupling of ECRA with biomass-derived substrates underscores the overall sustainability potential of this synthesis approach. However, current ECRA systems are limited by low substrate conversion, poor Faradaic efficiencies, competing side reactions, and high overpotentials. In this work, we overcome these limitations by developing a highly efficient electrocatalytic system for the reductive amination of biomass-derived levulinic acid as a model reaction. To this end, we use a simple two-electrode setup operated under galvanostatic conditions combined with the addition of ppm-amounts of lead as a catalytic additive. We achieve complete substrate conversion, Faradaic efficiencies of up to 70%, and product selectivities exceeding 98%. These findings demonstrate the potential of ECRA as a practical approach for efficient and sustainable primary amine synthesis.

1 | Introduction

Amines are important chemical compounds due to their diverse reactivity, biological relevance, and broad applicability across multiple scientific and industrial fields. For example, they are used in the synthesis of pharmaceuticals [1, 2], polymers [3], and in the agrochemical sector, where they can function as biocidal agents[4, 5]. Various synthetic methods exist for amine formation, including hydrogenation of nitro compounds, nitriles, and alkynes [6–8], C—H aminations [9–12], amination of alcohols [13], and reductive aminations [14–18]. Among these, catalytic reductive amination is a common strategy to introduce nitrogen-containing functionalities into a broad range of substrates. In these reactions, carbonyl compounds, for example, aldehydes and ketones, react with amines or ammonia in the presence of a catalyst and a reducing agent or cofactor [14–18]. The overall process generally proceeds in two distinct steps. First, the carbonyl compound undergoes a condensation reaction with the amine, resulting in the formation of a primary or secondary imine, or, in the case of hydroxylamine, the corresponding oxime (Scheme 1). In the subsequent step, the intermediate is reduced in the presence of an electron and a proton source, generating the desired primary or secondary amine. Electrocatalytic reductive amination (ECRA) represents a sustainable alternative to conventional methods, as it eliminates the need for stoichiometric molecular reducing agents or cofactors. Instead, electrons serve as cheap, clean, and efficient reducing agents [19, 20]. Furthermore, water can function simultaneously as both the proton source and the solvent. This approach is atom-efficient,



SCHEME 1 | Electrocatalytic reductive amination of carbonyl compounds ($R_1 = H$, C_nH_{2n+1}). The reaction proceeds via oxime formation through condensation of the carbonyl compound with hydroxylamine, followed by electrocatalytic reduction to the corresponding primary amine.

reduces reliance on costly and potentially hazardous reducing agents, minimizes waste generation, and avoids the production and storage of molecular hydrogen [20–22]. Accordingly, the use of hydroxylamine or ammonia as nitrogen sources in electrocatalytic amination reactions presents a sustainable and atom-efficient strategy for the synthesis of primary amines. Compared to other sustainable and atom-efficient reductive amination strategies, such as enzymatically catalyzed reductive amination [17, 18, 23–27], the presented ECRA approach eliminates the need for downstream recycling of reaction components (e.g., cofactors), providing a clear advantage in terms of process sustainability. While biocatalytic systems benefit from mild conditions and high enantioselectivity [17, 25], they are inherently constrained by enzyme specificity, which limits the overall substrate scope [23, 27], and shows sensitivity to pH and temperature [26, 27]. Although substrate scope can be partially expanded through enzyme engineering [25], the overall flexibility of enzymatic approaches remains limited. In contrast, ECRA offers broad tunability through the choice of electrode material, electrolyte composition, and applied potential or current.

First studies on the electrocatalytic synthesis of primary amines via the reduction of oximes were reported by Lund et al. in 1959. The authors demonstrated the fundamental feasibility of the electrochemical reduction of a broad spectrum of oximes. They tested non-conjugated, α,β-unsaturated, and aromatic derivatives using a mercury electrode [28]. In 2019, Fukushima et al. demonstrated the application of electrocatalytic oxime reduction for the synthesis of various amino acids. Hydroxylamine was used as the nitrogen source for the reductive amination of biomass-derived α-keto acids using a TiO_2/Ti mesh electrode in H_2SO_4 at pH 0.19. The method yielded Faradaic efficiencies ($F_{eff.}$) ranging from 77% to 99%. Particularly high conversions were obtained in the reaction of pyruvic acid to alanine ($X = 89\%$) with a Faradaic efficiency of 77%; however, this required a relatively high applied potential of −2.8 V versus reversible hydrogen electrode (RHE) [29]. In addition to ketones, aldehydes were also investigated as substrates by Schiffer et al., who studied the ECRA of benzaldehyde with ammonia to form benzylamine. The study focused on the influence of various metal catalysts, including Fe, Co, Rh, Ni, Pd, Pt, Cu, Ag, Au and Zn, in ammonia-saturated MeOH. Among these, Ag exhibited the highest $F_{eff.}$ (>75%) toward primary amine formation, albeit at a relatively low substrate conversion of 16% [30]. In a recent study, Rooney et al. reported the development of an electrocatalytic system for the selective reduction of oximes in a phosphate-buffered aqueous electrolyte, using acetone as a model substrate. They demonstrated a two-step, one-pot electrochemical process for the conversion of acetone to isopropylamine. The process consists of oxime formation, using nitrite as the amination reagent, on a Pd cathode. Subsequently, the cathode is replaced with Pb/PbO, on which the oxime is electrochemically reduced to the corresponding amine, achieving amine yields of up to 85% at −1.1 V versus RHE, albeit with moderate overall $F_{eff.}$ of 50% [31].

In 2021, Mürtz et al. demonstrated the applicability of levulinic acid in ECRA reactions using methylamine as the amination reagent. The secondary amine 4-(methylamino)pentanoic acid was obtained with yields and Faradaic efficiencies of up to 60%. Furthermore, reaction conditions for the reductive amination were systematically studied, identifying copper and silver next to lead as promising cathode materials and KH_2PO_4 as the most suitable electrolyte [21]. Beyond this, recent studies have indicated that metal hydride species may serve as catalysts or mediators in electrocatalytic reduction reactions. In 2021, Lucas et al. proposed a possible involvement of PbH_2 via a 'parallel electromediation path' in the reduction of levulinic acid to 4-hydroxyvaleric acid at a Pb cathode. Initial evidence for the formation of PbH_2 was provided by a signal in in situ attenuated total reflectance fourier-transform infrared spectroscopy (ATR-FTIR) experiments, which indicated an active Pb species in solution [32, 33]. Based on these findings, Kümper et al. verified the positive impact of trace amounts of lead in imine reduction in basic media and proposed a mechanism for the electrocatalytic hydrogenation (ECH) of the imine involving PbH_2. They propose that the hydride is formed at the cathode and subsequently hydrogenates the substrate in solution. These insights highlight the potential for tuning reaction efficiency by introducing trace amounts of lead as a catalytic additive in electrocatalytic systems [34].

Electrocatalytic reductive amination represents a promising alternative to conventional approaches for the synthesis of primary amines. However, low substrate conversions, undesired side reactions, poor Faradaic efficiencies, and the requirement for high potentials are still challenges to be faced. Comparable

challenges are also observed in alternative electrochemical strategies for primary amine synthesis, such as the ECH of nitriles [35–38]. In this study, we circumvent these limitations and present a highly efficient electrocatalytic system for the synthesis of primary amines by using the reductive amination of biomass-derived levulinic acid as a model reaction. By tuning the reaction conditions and incorporating trace amounts of lead as a catalytic additive, we achieve high Faradaic efficiencies, full substrate conversion, and product selectivities exceeding 95% within short reaction times under galvanostatic conditions. Thereby, we extend the Pb-catalyzed reaction pathway from imine reduction in basic media to oxime reduction in acidic media.

2 | Results and Discussion

ECRA via an oxime intermediate was investigated using levulinic acid as the substrate, which represents a biomass-derived platform chemical that has been extensively studied in related electrocatalytic reduction reactions. To expand the range of accessible downstream products derived from amines, such as pharmaceuticals, agrochemicals, and polymers, hydroxylamine is employed as an amination reagent for the selective synthesis of primary amines. The initial reaction conditions employed in this study are shown in Scheme 2.

Figure 1 illustrates the effective conversion of levulinic acid and hydroxylamine in an acidic environment: 4-aminopentanoic acid was obtained with a yield of 68% (±2%) in KH_2PO_4/H_2SO_4 at a pH of 1.5. First, the influence of different acids on the conversion and yields of the primary amine and alcohol was investigated. Maintaining the pH but using only H_2SO_4 as electrolyte led to a comparable amine yield of 69% (±1%). Interestingly, lower amine formation (47% ±1%) and higher alcohol formation (11.0% ±0.2%) were observed in HCl compared to the other electrolytes. The lower $F_{eff.}$ of 57.6% (±0.2%) indicates a loss of electrons to the hydrogen evolution reaction (HER). One possible explanation is a blocking effect of chloride ions at the cathode surface, where they occupy active sites. It is well established that specifically adsorbing anions exert a pronounced influence on electrochemical processes [39, 40]. For instance, chloride has been reported to block iron cathodes more effectively than sulfate during the HER, resulting in greater suppression of HER in HCl compared to H_2SO_4 [41]. This observation is consistent with the findings of Conway et al., who demonstrated that the specific adsorption of anions on transition metal surfaces increases in the order $F^- < ClO_4^- < SO_4^{2-} < Cl^- < Br^- < I^-$, which correlates with the decreasing Gibbs energies of hydration at 25°C of these anions [40, 42]. Accordingly, ions with lower hydration energies, and thus weaker solvation, exhibit stronger adsorption at the electrode surface [43]. In the present reaction system, such adsorption effects may also inhibit the adsorption of oxime reduction intermediates more strongly in HCl than in H_2SO_4.

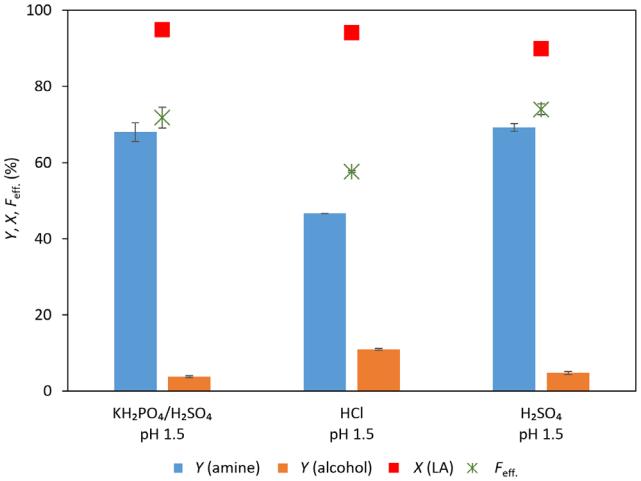
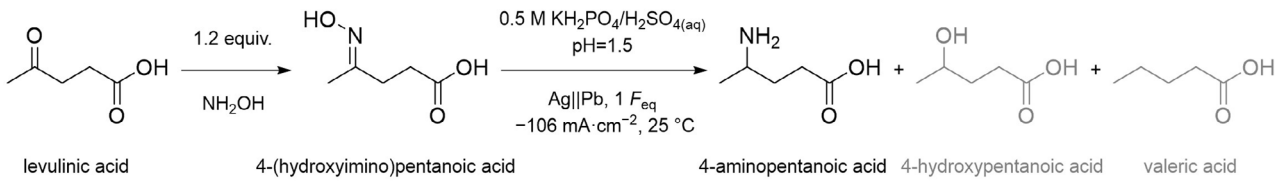


FIGURE 1 | Comparison of different electrolytes in the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid (Y (amine)) and 4-hydroxypentanoic acid (Y (alcohol)), as well as the conversion of levulinic acid (X (LA), and the $F_{eff.}$ are shown. $F_{eff.}$ takes into account the sum of electrons transferred to the amine and the alcohol during the ECH of LA. Reaction conditions: WE: mirror-like silver electrode; CE: Pb; $j = -212$ mA cm^{-2}; $F_{eq} = 1$; substrates: levulinic acid (LA): 2 M, hydroxylamine: 2.4 M; $T = 25$°C; pH 1.5. Experiments in KH_2PO_4/H_2SO_4 were carried out with a reaction solution volume of $V = 3$ mL, while $V = 2$ mL was used in the case of HCl and H_2SO_4.

2.1 | Influence of Pb on the ECRA of Levulinic Acid

Recently, Kümper et al. discovered a catalytic effect of traces of lead in reductive amination [34]. Because Pb anodes were used in our initial experiments, potential Pb leaching had to be considered. Reference experiments confirmed this effect, with ICP analysis detecting 4.7 ppm (±1.9%) Pb in the catholyte after electrolysis. In this context, PbH_2 has already been considered in previous research as an active participant in the electrocatalytic reduction of ketones, including levulinic acid [32, 34]. Based on these literature reports, we adopted the hypothesis that PbH_2 acts as the active species in the present system and subsequently investigated whether the presence of Pb^{2+} ions in solution influences the oxime reduction. To this end, the reaction was initially performed under Pb-free conditions. The Pb anode, a potential source of Pb contamination, was replaced with a Pt electrode (Figure 2).



SCHEME 2 | The initial reaction conditions: 2 M levulinic acid (LA) and 2.4 M hydroxylamine. The pH was adjusted by adding H_2SO_4 to a 0.5 M KH_2PO_4 solution, resulting in a pH of 1.5 at 25°C. The cathode and anode compartments were separated by an N-324 membrane. The anode compartment contained a 25% H_3PO_4 solution.

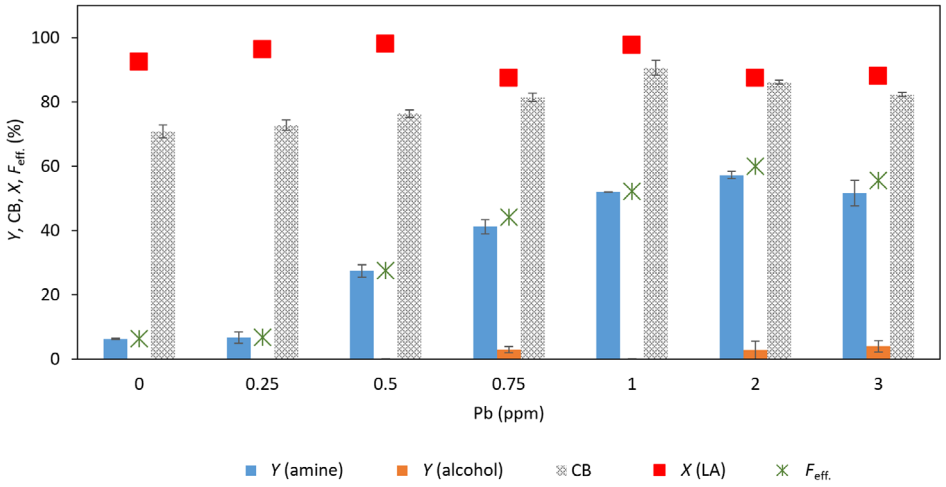


FIGURE 2 | Impact of different Pb concentrations on the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid *Y* (amine), 4-hydroxypentanoic acid *Y* (alcohol), the carbon balance (CB), the conversion of levulinic acid *X* (LA), as well as the $F_{eff.}$ are shown. Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -212\ mA \cdot cm^{-2}$; $F_{eq} = 1$; solvent: $KH_2PO_4/H_2SO_{4(aq.)}$ pH = 1.5; substrates: levulinic acid (LA): 2 M, hydroxylamine: 2.4 M; $T = 25°C$. $F_{eff.}$ takes into account the sum of electrons transferred to the amine and the alcohol during ECH of LA.

Under Pb-free conditions, the amine yield decreased to 6.3% (±0.2%), and no alcohol formation was observed (Figure 2). Subsequently, aqueous $Pb(NO_3)_2$ was added to the reaction solution in different concentrations. At a Pb concentration of 0.5 ppm, amine formation increased to 27% (±2%) (*X* (LA) = 98% (±2%)), indicating that the presence of Pb in the system is necessary for efficient amine production. Further increases in Pb concentration beyond 0.75 ppm resulted in elevated amine yields ranging from 41% (±1%) to 57% (±2%). On average, this corresponds to a maximum amine yield of approximately 49% at Pb concentrations exceeding 0.75 ppm. In addition to oxime reduction, elevated Pb concentrations also facilitate the direct reduction of LA to 4-hydroxypentanoic acid (*Y* (alcohol) = 2.9% (±1.0%) - 4.0% (±1.7%) at Pb > 0.75 ppm). These results indicate that the presence of Pb plays an essential role in the ECH of the oxime and LA. Based on the results presented in Figures 1 and 2, the Pb concentration series was repeated using sulfuric acid to examine whether the electrolyte type influences the oxime reduction in the presence of Pb. For the subsequent experiments (Figure 3), reaction conditions were optimized: the pH value of the reaction was set to 0, the equivalent charge (F_{eq}) was increased to 1.2 to ensure a slight excess of electrons, and the molar ratio of levulinic acid (LA) to hydroxylamine (NH_2OH) was adjusted to 1:1.6 to promote complete conversion of LA to the corresponding oxime (Figure S1). Additionally, the current density was reduced to –106 $mA \cdot cm^{-2}$ (Figure S2). Under Pb-free conditions, the amine yield decreased to 4.3% (±0.1%), and alcohol with a yield of 2.0% (±0.1%) was formed (CB = 80% (±3%)). Upon increasing the Pb concentration to 0.25 ppm, the amine yield increased significantly to 80% (±4%) with complete conversion of LA. A further increase in Pb concentration did not enhance amine formation; instead, it resulted in a higher yield of the by-product valeric acid (VA), which doubled from 1.5% (±0.2%) at 0.25 ppm Pb to 3.2% (±0.4%) at 2 ppm. These observations suggest that trace amounts of Pb ions generally promote ECH pathways over the HER under the given conditions. This trend is further underlined by the Faradaic efficiency, which increased from 5.2% (±0.1%) in the absence of Pb to 68% (±4%) upon Pb addition. In addition, the significantly enhanced amine formation at 0.25 ppm Pb in H_2SO_4 compared to a KH_2PO_4/H_2SO_4 mixture, from 7% (±2%) in KH_2PO_4/H_2SO_4 to 80% (±4%) in H_2SO_4, highlights the critical influence of the electrolyte composition on the effectiveness of Pb in the reaction system. Interestingly, the nature of the by-products depends on the presence of Pb. In its absence, the only by-product reaction observed was the two-electron reduction of the ketone to alcohol. In contrast, when Pb was present, VA was the only by-product detected. Comparable phenomena have been reported in the electrochemical reduction of levulinic acid under similar reaction conditions. Using a Pb cathode in 0.5 M H_2SO_4 at pH 0, Xin et al. observed a selectivity of 81.5%–97% for VA over GVL at 20% conversion of LA at an applied potential range of −1.1 to −1.5 V versus RHE [44]. Similarly, dos Santos et al. reported a VA selectivity of approximately 80% under an applied potential of –1.5 V versus standard hydrogen electrode (SHE) in 0.5 M H_2SO_4 [45]. In the interest of environmental safety, we eliminated the need for a Pb electrode and instead introduced a controlled concentration of Pb ions. This approach enabled us to reduce the dissolved Pb concentration to 0.25 ppm, which is significantly lower than levels observed after electrode leaching (4.7 ppm (±1.9%)), while maintaining high catalytic performance.

2.2 | Identification of Factors Influencing the Efficiency of Pb in Oxime Reduction

If PbH_2 is considered the active species in the observed results [34], its formation and stability, and therefore its reactivity, are likely influenced by the nature of the electrolyte. The literature on the electrochemical synthesis of metal hydrides discussed electrolyte composition, acidity, and pH as critical factors [33, 46–48]. For example, Denkhaus et al. observed that hydride formation efficiency increases with higher acid concentrations. They also found that, of the electrolytes tested, H_2SO_4 showed the best performance in the formation of arsine and stibine [47]. Additionally, Arbab-Zavar et al. observed a similar trend in the formation of thallium

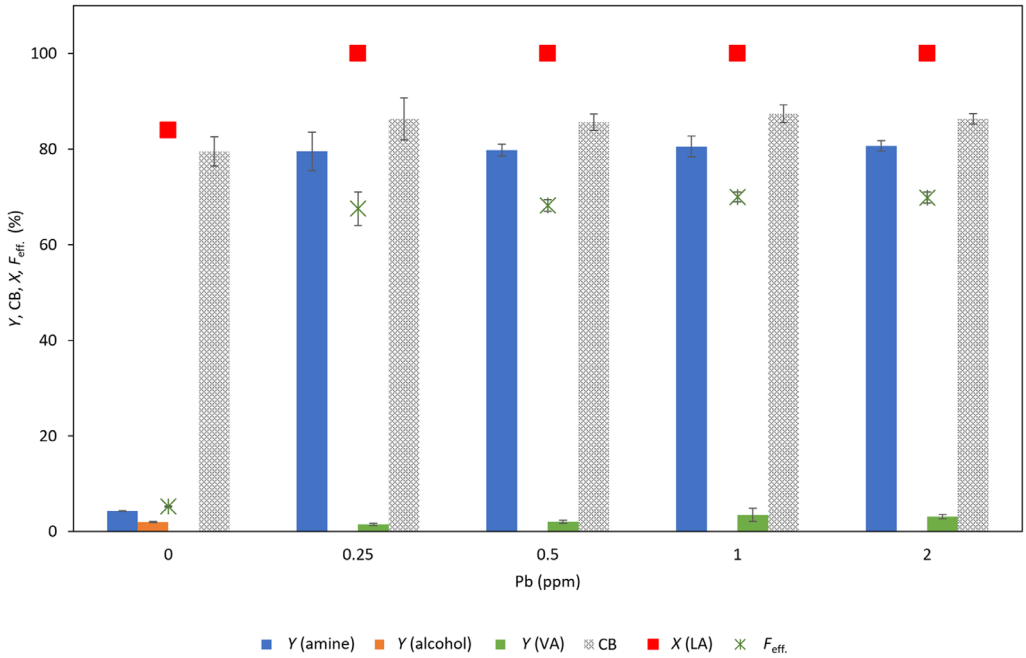


FIGURE 3 | Impact of different Pb concentrations on the reductive amination of levulinic acid with hydroxylamine under optimized reaction conditions: H_2SO_4 electrolyte, pH = 0. The yields of 4-aminopentanoic acid *Y* (amine), 4-hydroxypentanoic acid *Y* (alcohol), valeric acid *Y* (VA), the conversion of levulinic acid *X* (LA), the carbon balance (CB), as well as the Faradaic efficiency ($F_{eff.}$) are shown. Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -106\ mA \cdot cm^{-2}$; $F_{eq} = 1.2$; solvent: H_2SO_4 pH = 0; substrates: levulinic acid (LA): 2 M, hydroxylamine: 3.2 M; $T = 25°C$. $F_{eff.}$ takes account of the sum of electrons transferred to the amine, alcohol, and VA during ECH of LA.

hydride species [48]. As shown in Figure 1, in the present study, the selected electrolyte was also found to influence the reduction of oximes. Moreover, the electrolyte composition appears to affect Pb efficiency in the reaction system (Figures 2 and 3). Given the pronounced effect of Pb^{2+} ions on oxime reduction, presumably via the formation of an active Pb hydride species, the influence of the electrolyte on Pb efficiency in the current reaction system was investigated further (Figure 3). To maintain a low pH for oxime reduction, various acids were investigated as electrolytes, and three inexpensive, readily available acids of differing acidity were selected (Table 1).

Figure 4 shows that, in agreement with the observations in H_2SO_4 and KH_2PO_4/H_2SO_4 systems, negligible formation of the amine occurs in the absence of Pb. Upon addition of 0.25 ppm Pb, a significant increase in amine formation was observed in all acids, with notable differences in amine formation among the different acids. The highest amine yield was achieved in H_2SO_4, followed by H_3PO_4, HCl, and HNO_3. Further increases in Pb concentration resulted in enhanced amine formation in HCl and HNO_3, reaching maximum yields of 70% (±1%) in HCl, and 32% (±4%) in HNO_3 at 5 ppm Pb. This trend suggests that amine formation could be further improved by increasing Pb concentration. In case of H_2SO_4 and H_3PO_4, a plateau is reached at concentrations above 0.25 ppm Pb, reaching amine yields of around 80% and 75%, respectively. Minor alcohol formation was detected in all acids only under Pb-free conditions. In contrast to H_2SO_4 (Figure 3), no formation of VA was observed in

TABLE 1 | Tested electrolytes and their pKa [49].

Acid	pKa
HCl	−2
H_2SO_4	−2
HNO_3	−1.32
H_3PO_4	2.16

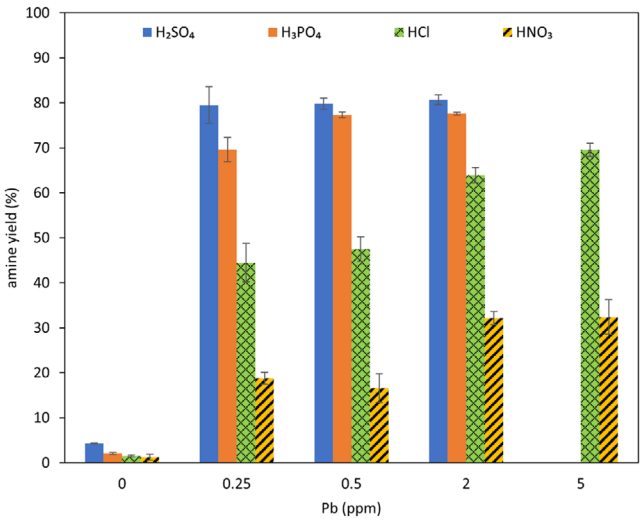


FIGURE 4 | Influence of the acid on the efficiency of Pb in the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid are presented. Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -106\ mA \cdot cm^{-2}$; $F_{eq} = 1.2$; solvent: HCl, H_2SO_4, HNO_3, H_3PO_4 pH = 0; substrates: levulinic acid (LA): 2 M, hydroxylamine: 3.2 M; $T = 25°C$; anolyte: 25% H_3PO_4. The experiments shown were carried out in triplicate for H_2SO_4 and HNO_3 and in duplicate for HCl and H_3PO_4.

any of the other acids. These results indicate no correlation between acid strength and the reduction efficiency of Pb.

Overall, the trend observed in Figure 4 correlates with the observed trend in Figure 1, whereby amine formation in HCl is significantly less pronounced than in sulfuric acid. As mentioned above, this trend can possibly be attributed to a blocking effect caused by the adsorption of the corresponding anions on the electrode surface. In this case, the Gibbs energy of ion hydration can provide information about the tendency of the respective ions to absorb on the electrode. When the Gibbs energy of ion hydration for the anions of the tested electrolytes is considered as an indicator, the following trend emerges: $HPO_4^{2-} < SO_4^{2-} < H_2PO_4^- < HSO_4^- < Cl^- < NO_3^-$ [42]. This corresponds to the trend observed in Figure 4 and supports the hypothesis that the differing efficiencies of Pb in oxime reduction are caused by a blocking effect induced by anion adsorption.

Since the pH is known to be a critical factor for the formation, stability, and reactivity of PbH_2, in addition to the acidity of the electrolyte, the reaction system was also optimized with regard to the pH [33, 46–48]. Therefore, the dependence of oxime reduction on pH in the presence of Pb was investigated using H_2SO_4 as the electrolyte at a Pb concentration of 2 ppm (Figure 5). Increasing the pH resulted in a pronounced decrease in amine formation, from 81% (±1%) at pH 0 to 30% (±4%) at pH 3.5 and further down to 24% (±2%) at pH 7. Complete conversion of LA was observed across the studied pH range. However, oxime reduction was progressively inhibited with increasing pH as the residual oxime concentration after electrolysis rises with increasing pH from 0% at pH 0 to 49% (±7%) at pH 3.5 and 73% (±9%) at pH 7. This effect may arise from the fact that the protonated, positively charged oxime molecule is known to be reduced at less negative potentials than the neutral species present in neutral to basic solutions [50]. At higher pH, the requirement for more negative reduction potentials favors HER over oxime reduction. An additional contributing factor could be a pH-dependent formation of PbH_2 [33, 46–48].

To quantify the catalytic efficiency of Pb in the hydrogenation reaction across different electrolytes, the turnover number (TON) provides a meaningful measure. It refers to the number of substrate molecules converted into product per catalyst molecule [51, 52]. Based on the hypothesis that the PbH_2 species acts homogenously as a mediator or catalyst [34], we employ the standard practice for homogenous catalytic systems and assume that all Pb ions present in solution participate in the reaction. This assumption is further supported by the exclusion of a heterogeneous pathway involving Pb deposition on the Ag electrode (see Figure S4). Figure 6 illustrates the comparison at Pb concentrations of 0.25 and 2 ppm for each acid. Under the conditions with 0.25 ppm Pb, H_2SO_4 exhibits the highest efficiency, with a TON of 1.31 (±0.07)·10^6, significantly outperforming all other systems. A comparable TON is observed only in H_3PO_4 (1.18 (±0.05)·10^6). In contrast, using HCl yields a considerably lower TON of 0.73 (±0.07)·10^6, approximately half that obtained in H_2SO_4. Utilizing HNO_3 results in a TON of 0.31 (±0.02)·10^6. In comparison, homogeneous Rh-catalyzed reductive amination of benzaldehyde with ammonia lead to TONs of only up to 1720 [53]. Additional TONs for Ru-catalyzed reductive amination of various substrates were reported by Fatkulin et al., ranging from 1020 to 8875 [54]. The results of the present study not only underline the pronounced effect of the electrolyte on the efficiency of Pb but also illustrate the remarkable efficiency of Pb-catalyzed ECRA reactions compared to non-electrochemical homogeneously catalyzed methods.

Under these conditions, the system's overall efficiency could be increased further (Figure 7). When the current density was increased from −106 to −212 and −318 mA cm^{-2}, the amine yield remained largely constant at 80% (±4%), 76% (±2%), and 77% (±5%), respectively. In this context, the turnover frequency

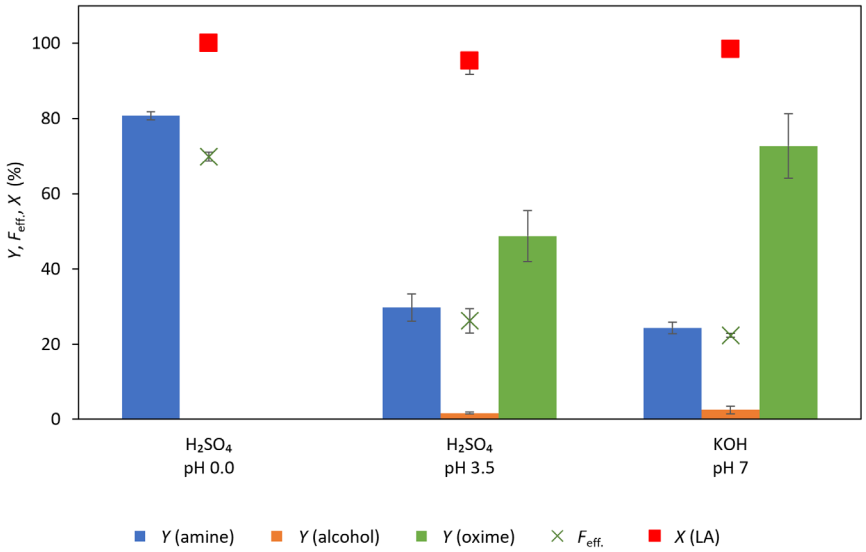


FIGURE 5 | Impact of different pH in the presence of Pb on the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid *Y* (amine), 4-hydroxypentanoic acid *Y* (alcohol), 4-(hydroxyamino)pentanoic acid, *Y* (oxime), as well as the Faradaic efficiency ($F_{eff.}$) are shown. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -106$ mA cm^{-2}; $F_{eq} = 1.2$; solvent: H_2SO_4 pH = 0, 3.5, and 7; substrates: levulinic acid (LA): 2 M, hydroxylamine: 3.2 M; $Pb(NO_3)_2$: 2 ppm; $T = 25$°C. $F_{eff.}$ takes into account the sum of electrons transferred to the amine and alcohol during ECH of LA. The experiments shown were carried out in duplicates.

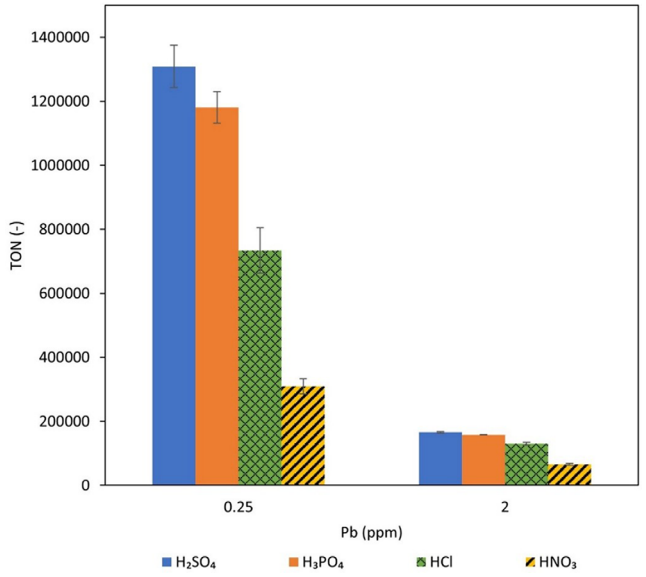


FIGURE 6 | Influence of the acid on the efficiency of Pb in the reductive amination of levulinic acid with hydroxylamine. TONs are presented at different Pb concentrations: 0.25 ppm and 2 ppm. Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -106\ mA \cdot cm^{-2}$; $F_{eq} = 1.2$; solvent: HCl, H_2SO_4, HNO_3, H_3PO_4 pH = 0; substrates: levulinic acid (LA): 2 M, hydroxylamine: 3.2 M; T = 25°C; anolyte: 25% H_3PO_4. The experiments shown were carried out in triplicate for H_2SO_4 and HNO_3 and in duplicate for HCl and H_3PO_4.

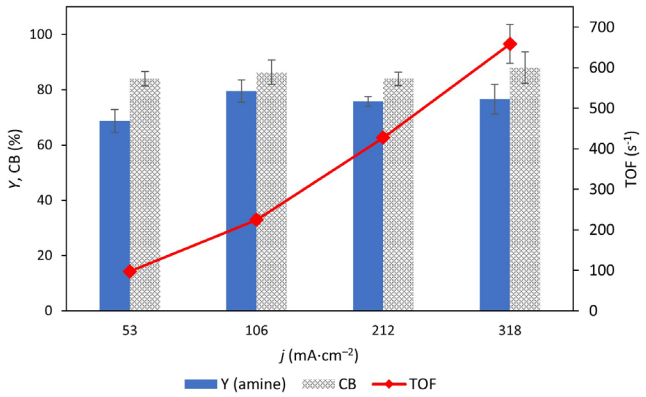


FIGURE 7 | Influence of the current density j on amine formation and the efficiency of Pb in the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid Y (amine), the carbon balance (CB), and the TOFs are presented. 0.25 ppm Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -53, -106, -212, -318\ mA \cdot cm^{-2}$; $F_{eq} = 1.2$; solvent: H_2SO_4 pH = 0; substrates: levulinic acid (LA): 2 M, hydroxylamine: 3.2 M; T = 25°C; anolyte: 25% H_3PO_4. The experiments shown were carried out in triplicate.

(TOF) is a useful measure of Pb activity as it describes the reaction rate by relating the TON to time [52]. Consequently, the TOF increased with current densities ($225\ s^{-1}$ (±11), $427\ s^{-1}$ (±10), and $658\ s^{-1}$ (±48)), indicating higher Pb efficiency without compromising product yield. However, decreasing the current density to 53 mA cm^{-2} led to a decline in the amine yield to 69% (±4%) and to a decrease in TOF to 97 (±6.3). In comparison, Chandra et al. showed TOFs of up to $1850\ hr^{-1}$ (= $0.51\ s^{-1}$) for the production of primary amines by reductive amination of various carbonyl compounds with Rh nanoparticles [55].

3 | Conclusion

In summary, we have developed a highly efficient system for the synthesis of primary amines via ECRA of ketones, using the conversion of levulinic acid with hydroxylamine to 4-aminopentanoic acid as a model reaction. Initial investigations showed that, with Ag as cathode and Pb as anode, the choice of electrolyte strongly affects the reaction performance, likely due to a cathode surface blocking effect by the electrolyte anions that varies with the respective anions. Furthermore, the presence of Pb was shown to be crucial for the reduction of the oxime intermediate, with PbH_2 assumed to be the active species. Accordingly, the addition of 0.25 ppm Pb increased the yield of the primary amine from 4% in the absence of Pb to 80%, while achieving a selectivity of 98%. Thereby, we successfully extended the Pb-catalyzed reaction pathway from imine reduction in basic media to oxime reduction in acidic media. Furthermore, it was found that the efficiency of Pb in oxime reduction depends on the chosen electrolyte. In this context, as a measure of efficiency, the TON was employed, which increases in the following order: HNO_3 < HCl < H_3PO_4 < H_2SO_4. Using H_2SO_4, a maximum TON of $1.31\ (\pm 0.07) \cdot 10^6$ was obtained. This value exceeds the TONs of Ru-catalyzed reductive amination reactions by approximately 150-fold, highlighting the extraordinary efficiency of Pb-catalyzed ECRA reactions compared to homogeneously catalyzed methods. The efficiency of the reaction system could be further enhanced by increasing the current density, resulting in Pb TOFs of up to $658\ s^{-1}$. Overall, the presented reaction system not only stands out by its high conversion, product yields, and selectivity, but also by its operational simplicity: mild reaction conditions, two-electrode setup, galvanostatic conditions, and minimal catalyst loadings providing a foundation for further scale-up and development.

Acknowledgments

The authors thank the Federal Ministry of Food and Agriculture for funding within PyrSElekt (FNR 2220NR101X). The authors acknowledge the Cluster of Excellence Fuel Science Center (EXC 2186, ID: 390919832) by the Excellence Initiative of the German federal and state governments to promote science and research at German universities for funding. In addition, JK and RP acknowledge funding by the German Research Foundation (ElectroPyr, 459861727). Philippe Sautet, Yani Guan, and Simran Kumari were supported by the National Science Foundation grant 2140374. Moreover, the authors thank Philipp Baginski and Leander Brück for their work in the lab and for performing some of the discussed experiments. Additionally, the authors thank Ines Bachmann-Remy and Dr. Meike Emondts for the NMR measurements and Heike Bergstein for the ICP measurements. N. K. thanks the Leopoldina Fellowship Program, National Academy of Sciences, Leopoldina grant number LPDS 2023–08.

Open Access funding enabled and organized by Projekt DEAL.

Funding

This study was supported by Fachagentur Nachwachsende Rohstoffe (Grant FNR 2220NR101X), Deutsche Forschungsgemeinschaft (Grant

EXC 2186, ID: 390919832, 459861727), National Science Foundation (Grant 2140374), Deutsche Akademie der Naturforscher Leopoldina - Nationale Akademie der Wissenschaften (Grant LPDS 2023-08).

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting information section. **Supporting Fig. S1:** Influence of the ratio NH_2OH to levulinic acid (LA) on LA conversion *X* (LA). The ^{1}H NMR spectrum of the reaction solution prior to electrolysis containing NH_2OH (1, 2, 3 and 4 M) and LA (2 M) in KH_2PO_4/H_2SO_4. Levulinic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 2.05 (s, 3H; CH_3), 2.29-2.44 (m, 2H; CH_2), 2.62 (t, $^3J_{H,H}$ = 6.3 Hz, 2H; CH_2), cis-4-(hydroxyimino)pentanoic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.76 (s, 3H; CH_3), 2.29-2.44 (m, 4H; CH_2); trans-4-(hydroxyimino)pentanoic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.70 (s, 3H; CH_3), 2.29-2.44 (m, 4H; CH_2); 1,3,5-Trioxane (STD): ^{1}H NMR (400 MHz, DMSO-d_6): δ = 5.05 (s, 6H; CH_2). **Supporting Fig. S2:** Influence of the current density *j* on amine formation in the reductive amination of levulinic acid with hydroxylamine. The yields of 4-aminopentanoic acid *Y* (amine), the carbon balance (CB), and levulinic acid conversion *X* (LA) are presented. Reaction conditions: WE: mirror-like silver electrode; CE: Pb; j = –42, –106, –304 mA cm^{-2}; F_{eq} = 0.5; solvent: KH_2PO_4/$H_2SO_{4(aq.)}$ pH = 1.5; substrates: levulinic acid (LA): 2 M, hydroxylamine: 2.4 M; T = 25 °C; anolyte: 25% H_3PO_4. The experiments shown were carried out in duplicate. **Supporting Fig. S3:** ^{1}H NMR spectrum of the catholyte solution after electrolysis of levulinic acid with hydroxylamine in H_2SO_4. Levulinic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 2.04 (s, 3H; CH_3), 2.25-2.41 (m, 2H; CH_2), 2.61 (t, $^3J_{H,H}$=6.3 Hz, 2H; CH_2), cis-4-(hydroxyimino)pentanoic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.72 (s, 3H; CH_3), 2.25-2.41 (m, 4H; CH_2); trans-4-(hydroxyimino)pentanoic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.68 (s, 3H; CH_3), 2.25-2.41 (m, 4H; CH_2); 7.69 (s, 1H; OH) 4-aminopentanoic acid: ^{1}H NMR (400 MHz, DMSO-d_6): δ =1.12 (d, $^3J_{H,H}$ = 6.5 Hz, 3H; CH_3), 1.56-1.66 (m, 1H; CH_2), 1.74-1.85 (m, 1H; CH_2), 3.09-3.22 (m, 1H; CH) 1,3,5-Trioxane (STD): ^{1}H NMR (400 MHz, DMSO-d_6): δ = 6.21 (s, 6H; CH_2). **Supporting Fig. S4:** Test of possible deposition of Pb on the Ag electrode. The yields of 4-aminopentanoic acid *Y* (amine) and 4-(hydroxyimino)pentanoic acid *Y* (oxime) as well as the conversion of levulinic acid *X* (LA) are shown. Pb was added as aqueous $Pb(NO_3)_2$. Reaction conditions: WE: mirror-like silver electrode; CE: Pt; $j = -106$ mA · cm^{-2}; F_{eq} = 1.2; solvent: H_2SO_4 pH 0; substrates: levulinic acid (LA): 2 M, hydroxylamine: 2.4 M; T = 25 °C. The same Ag electrode was reused for all three experiments. In the 1st run, a freshly polished Ag electrode was used under Pb-free conditions for reference. After rinsing and drying (without polishing), the Ag electrode was reused in the 2nd run, where 2 ppm Pb was added. The Ag electrode was then rinsed and dried again and reused in the 3rd run under Pb-free conditions to assess whether Pb deposition had occurred during the 2nd run. **Supporting Fig. S5:** Illustration of the specially developed electrochemical set-up used for the electrolysis experiments including an electrochemical cell consisting of three cuboid modules stacked on top of each other, a power supply, and a cooling system. **Supporting Table S1**. Overview of the reaction parameters, product yields and faradaic efficiencies for the ECRA of levulinic acid with hydroxylamine.