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Electrochemical hydrogenation for water purification made easy

Emmanuel Mousset

► **To cite this version:**

Emmanuel Mousset. Electrochemical hydrogenation for water purification made easy. *Nature Water*, 2023, 1 (1), pp.28-29. 10.1038/s44221-022-00011-2 . hal-04236946

HAL Id: hal-04236946

<https://hal.science/hal-04236946v1>

Submitted on 11 Oct 2023

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1 **WASTEWATER TREATMENT**

2 **Electrochemical hydrogenation for water purification made easy**

3 *The design of an electrochemical system without electrolytes allows hydrogenation of oxidized*
4 *contaminants in water without the introduction of additional chemicals*

5 Emmanuel Mousset

6 *Université de Lorraine, CNRS, LRGP, F-54000 Nancy, France*

7 email: emmanuel.mousset@cnrs.fr

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9 The rise of water demand through agriculture, industries and domestic activities, combined with the
10 water stress that is associated with climate change, is already leading to increased usage conflicts [1].
11 The water crisis produces by the increased demand and reduced availability of clean water represents
12 one of the biggest challenges of the 21st century and is part of the sustainable development goal
13 addressed by the United Nations [1].

14 Among the various approaches to temper the problems created by water scarcity, the reuse of the
15 treated wastewater for agriculture (e.g. irrigation), for industry or even for drinking water is a
16 palatable solution [1,2]. However, most of the wastewater treatment plants (WWTPs) currently in use
17 are based on biological processes and are not designed to completely eliminate recalcitrant pollutants
18 such as pesticides, pharmaceuticals and cosmetics. These contaminants are found at trace levels (ng/L
19 to µg/L) in natural water, and even at such ranges of concentration they can pose substantial
20 environmental and health risks [3].

21 Physico-chemical treatments for wastewater could be an attractive alternative to biological process
22 to remove persistent pollutants [4]. Before they can widely employ however, these technologies
23 advance to the stage where they require low cost and they provide high level of efficiency.
24 Furthermore, it is essential that they operate without the substantial need of chemicals that can pose
25 additional environmental risks to the ones they are intended to reduce, and research is currently in
26 progress to achieve these conditions. Now writing in Nature Water, Yinghua Xu et al. report an
27 electrochemical system that could be used to treat wastewater without the substantial introduction of
28 chemical that could become water contaminants [5].

29 Electrochemical systems are arising growing interest, because only electrons (ne^-) are involved to
30 eliminate the water pollution [6]. This can be done in two main ways. The first possibility is that the
31 pollutants can be electro-oxidized or electro-reduced with a direct electron transfer at the anode or

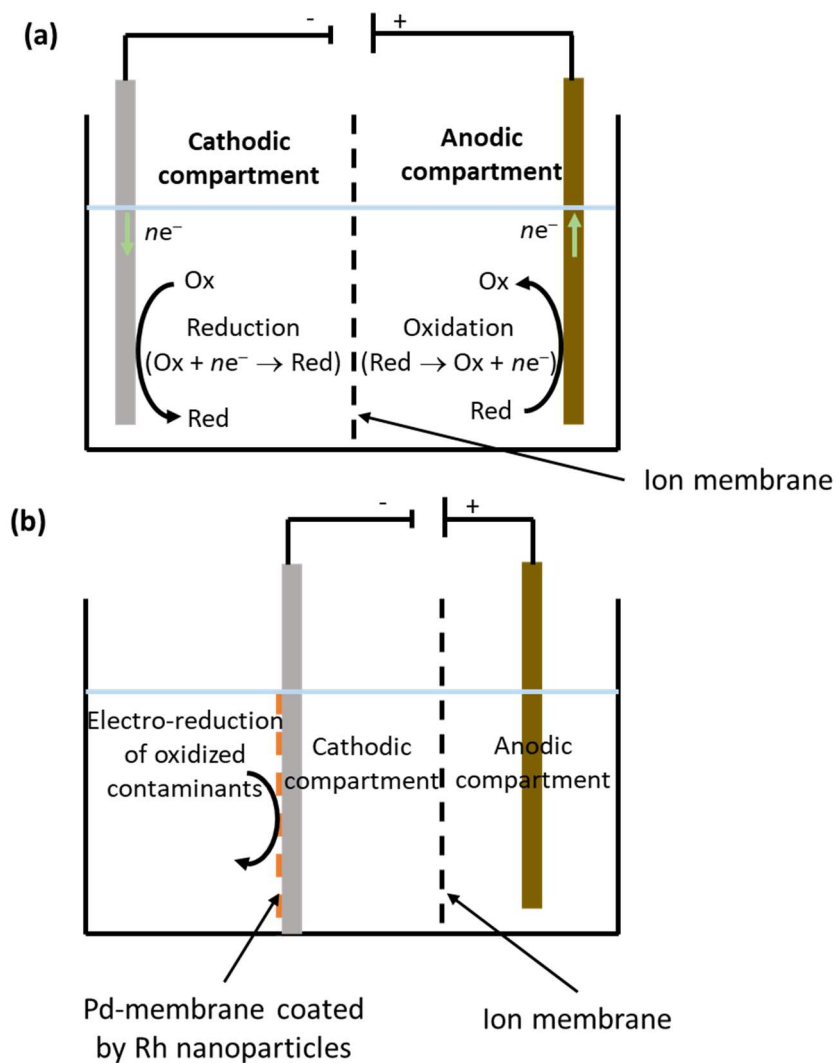
32 cathode surface, respectively (Figure 1a). The second path is the electro-oxidation or electro-
33 reduction of the solvent (H₂O in this case) and/or of other ionic species (e.g., SO₄²⁻, Cl⁻) that can then
34 form strong oxidants (e.g., hydroxyl radicals ([•]OH), sulfate radical (SO₄^{•-}), chlorine radical (Cl[•]))
35 and/or reductants (aqueous electron (e⁻_(aq))) that will eliminate the water pollution. This is called the
36 indirect oxidation or reduction. Electrochemical technologies can also be flexible, easily scalable and
37 allow operating numerous separation and conversion systems [7]. According to the operating
38 conditions (e.g. applied current density, reactor design, type of electrode materials, water
39 characteristics), high oxidation or reduction power can be obtained. This can lead to very high
40 conversion rates and yields (>99%), as defined by the difference of initial concentration of the
41 compounds to be removed with concentration at time *t*, divided by the initial concentration. However,
42 the solution to treat need to be conductive enough to power the electrolysis. In many effluents, and
43 particularly in municipal WWTPs, the ion content is not high enough so that the addition of
44 supporting electrolyte is required. Moreover, the oxidative treatments often produce unwanted by-
45 products (e.g. chlorate, perchlorate, nitrate) in presence of chloride ions (Cl⁻) and/or nitrogenous
46 species that are ubiquitous in water. This problematic with intermediates is inherent with all
47 conversion systems. In addition, some of the organic pollutants are at too high oxidation state (e.g.
48 haloacetic acid, chlorophenol), meaning that a reduction technique is preferable [8].

49 Xu et al. propose to avoid the addition of a supporting electrolyte by implementing a divided cell in
50 which the electrochemical reactions occur behind the electrode, rather than in between cathode and
51 anode like in a conventional reactor (Figure 1b) [5]. They also used palladium (Pd) membrane
52 modified with rhodium (Rh) nanoparticles placed in the cathode chamber to catalyze the reduction of
53 oxyanions (chlorate, perchlorate, bromate, nitrite, nitrate), haloacetic acids and chlorophenols by
54 hydrogenation. They succeeded to reach conversions yield higher than 99%, except with nitrate and
55 perchlorate. The high selectivity towards acetic acid is attributed to the Rh nanoparticles that favor

56 their adsorption compared to Pd, as noticed with Density Functional Theory calculation. The
57 performance is confirmed in real tap water with around 85% decrease of trichloroacetic acid, and lead
58 to a level of contamination below the threshold value (60 µg/L) set by the USEPA.

59 The results of Xu et al. are promising; however, some challenges remained to be solved. The lifetime
60 of this kind of membrane electrode has to be tested over a long period (e.g. several months). This
61 parameter is important, particularly in terms of costs, since it requires the use of expensive metals.
62 This will be a prohibitive solution at larger scale if the assays failed. Moreover, scale up studies will
63 have to be performed to further assess the efficiency at pilot scale. Furthermore, the cathode scaling
64 is often encountered with solution that contain calcium and carbonates, which is the case of numerous
65 effluents. So, the polarity reversal technique will need to be tested with such electrode materials,
66 otherwise passivation issues will be encountered in continuous mode and at large scale. Finally, the
67 control of selectivity in the aim at producing reduced valuable compounds could even be of higher
68 interest.

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 71 **Figure 1.** (a) schematized conventional divided electrochemical reactor (Ox: oxidant, Red:
 72 reductant, e^- : electron, n : number of electrons transferred), (b) schematized reactor design proposed
 73 by Xu et al. to operate without supporting electrolyte and for the electro-reduction of oxidized
 74 organic (haloacetic acids and chlorophenols) and inorganic (chlorate, perchlorate, bromate, nitrite,
 75 nitrate) contaminants in water [5].

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107 The authors declare no competing interests.

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