

Model-based control design for H₂ purity regulation in high-pressure alkaline electrolyzers

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Abstract

This paper proposes two control strategies that mitigate the cross contamination of H₂ and O₂ in a high-pressure alkaline electrolyzer, which consequently increases the supplied gases purity: one based on a decoupled PI scheme and the other based on optimal control tools. In order to reduce the diffusion of gases through the membrane, the controllers establish the opening of two outlet valves based on the pressure of the system and the difference in liquid level between both separation chambers. Therefore, two multiple input - multiple output controllers are designed here. For this purpose, a high-fidelity model previously developed was simplified in order to obtain a control-oriented model. The proposed controllers were evaluated in simulation using the high-fidelity nonlinear model in a wide operating range, which resulted in less than 1% impurity of gases.

Keywords: Hydrogen, alkaline electrolysis, multivariable control, \mathcal{H}_∞ optimal control

1. Introduction

The world economy is constantly expanding along with the demand for energy [1]. Furthermore, the extensive use of fossil fuels, with the consequent

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4 emission of greenhouse gases, is widely accepted as a situation that needs to
5 change. In this line, global impact studies and environmental protection
6 policies have been formulated [2, 3]. Around the world, solutions focused
7 on renewable energy sources have been proposed in order to mitigate the
8 emission of greenhouse gases due to the intensive use of fossil fuels. However,
9 the ability to accumulate the excess of energy over long periods of time is
10 needed in order to reach a high integration of renewable energy sources. A
11 widely accepted idea is the use of hydrogen as an energy vector, known as
12 the *hydrogen economy*, which would be an integral solution to produce, store
13 and supply energy [4, 5, 6].

14 Among all the methods of producing sustainable hydrogen, the alkaline
15 electrolysis is presented as the most available technology. Currently, there is
16 a renewed interest in this technology due to its ease of connection to renew-
17 able energy sources [7]. Commonly, the combination of electrolyzers, storage
18 tanks and fuel cells is used as an energy buffer [8, 9]. Alkaline electrolysis
19 consists in the separation of water to form H_2 and O_2 by applying an electric
20 current. The electrolytic cell consists of a pair of electrodes and a mem-
21 brane made of ZirfonTM that prevent gas mixing. One of the most important
22 challenges of the alkaline electrolysis is the diffusion through this membrane
23 driven by differences in concentration and pressure [10]. Although the first
24 cause of cross-contamination is inherent in the process and is related to the
25 development of new membranes, the pressure differential can be mitigated by
26 a suitable control design actuating over the outlet valves of both separating
27 chambers.

28 Despite alkaline electrolysis is a mature technology, its mathematical
29 modelling is still under development. Most models focus only on the cell-
30 stack description but not in the entire system [11, 12, 13]. Moreover, most
31 of them describe the stationary regime and are built from empirical equa-
32 tions [14, 15, 16]. Recently, Sanchez et al [17] used a commercial software to
33 model the entire system while the cell-stack is described by a semi-empirical
34 approach. In the same direction, some of the authors of the current work
35 have developed a Phenomenological Based Semi-empirical Model (PBSM) re-
36 ported in [18, 19]. This model has the advantage of describing the dynamic
37 phenomena and the evolution of all the electrolyser subsystems.

38 Furthermore, to the best of the authors' knowledge, and also from the
39 conclusions reported by Olivier et al [20], the design of controllers to solve
40 the problem mentioned above seems to be not addressed yet in the literature.
41 Therefore, the development of useful input-output models for control design

42 is an open research topic [20]. In general, control objectives are completely
43 focused on the management of the electrolyzer as an electrical consumer
44 and producer of H_2 connected to a grid [21, 22]. Moreover, the control of
45 the outlet valves could be found mentioned only by Schug in his description
46 of a pilot plant [23]. In his work, an alkaline electrolyzer is described in
47 detail along with experimental results. However, the control system is not
48 detailed enough, but the connection of plant output with control action can
49 be recognized in the simplified flow diagram presented.

50 Given the lack of control strategies designed for such systems and, in
51 particular, those strategies based on suitable and reliable (dynamic) mod-
52 els properly obtained for control tasks, the main contribution of this paper
53 is twofold. First, from a well-established nonlinear model considering the
54 dynamics and the accurate phenomenology of the alkaline electrolyzers re-
55 ported in [19], a reduced model able to be used as a control-oriented model
56 (COM) is obtained and properly validated by using the complete nonlinear
57 model (which, in turn, is validated with real data). Second, by using the
58 reduced model, two controllers are designed and the closed-loop performance
59 of the system is compared based on the maximization of the hydrogen purity
60 through the mitigation of the cross-contamination of gases into the chambers.

61 The remainder of the work is structured as follows. A description of a
62 high-pressure alkaline electrolyzer is presented in Section 2. Next, in Sec-
63 tion 3, two controllers are designed, a multivariable PI controller and an
64 optimal model-based one. Simulation results comparing both controllers are
65 presented and discussed in Section 4. At the end, some final comments are
66 gathered in Section 5.

67 **2. High-pressure alkaline electrolyzer**

68 As previously mentioned, a proposed solution for energy storage is the
69 combination of an electrolyzer, storage tanks and a fuel cell. In this way, the
70 additional electrical energy is used to produce hydrogen that is stored in the
71 tanks. When renewable energy sources are not able to meet the demand, the
72 stored hydrogen is consumed by the fuel cell.

73 High-pressure alkaline electrolyzers can supply gases at a storage pressure,
74 dispensing with the use of compressors. However, cross-contamination, i.e.,
75 the concentration of O_2 in the H_2 stream and vice versa, increases with
76 pressure, then special attention is required in operation due to safety and
77 quality issues.

78 Figure 1 shows the piping and instrumentation of a high-pressure alkaline
79 electrolyzer prototype. The components of this system are:

- 80 • a pressurized tank (PT) that contains a pack of 15 alkaline electrolytic
81 cells;
- 82 • two independent KOH solution circuits with recirculation pumps;
- 83 • two gas separation chambers (SC) where the produced gas is split from
84 the liquid KOH solution;
- 85 • two heat exchangers for both circuits (HEO and HEH);
- 86 • a water injection pump that periodically replenishes the consumed wa-
87 ter;
- 88 • two outlet lines controlled by two motorized valves (MVO and MVH)
89 connected to storage tanks; and
- 90 • an equalization line that connects both bottoms of the SCs.

91 A detailed description of this system is presented in [18, 19].

92 As mentioned in the Introduction, the main objective of an alkaline elec-
93 trolyzer is to separate water to form H_2 and O_2 by applying an electric
94 current I . In this process, it is highly important to minimize the diffusion
95 through the membrane caused by differences in both concentration and pres-
96 sure. Up to 2% of H_2 in the O_2 stream is widely accepted as a limit, taking
97 into account that the lower explosive limit of H_2 is 4%. Additionally, H_2 and
98 O_2 gases must be delivered at high pressures in order to avoid the use of
99 compressors. Since gas purity decreases with higher pressures, it is expected
100 to increase the possible operating pressure preventing contamination with a
101 suitable control strategy.

102 2.1. Cross-contamination

103 As stated before, the main difficulty in the operation of an alkaline elec-
104 trolyzer is the contamination of both streams, especially on the O_2 side.
105 Generally, this concept is approached in the models as an empirical equation
106 that relates contamination to the state of the system (e.g., current density,
107 temperature, pressure). This way evidences the lack of dynamic analysis of
108 purity. However, there are studies that analyze the phenomenology of the

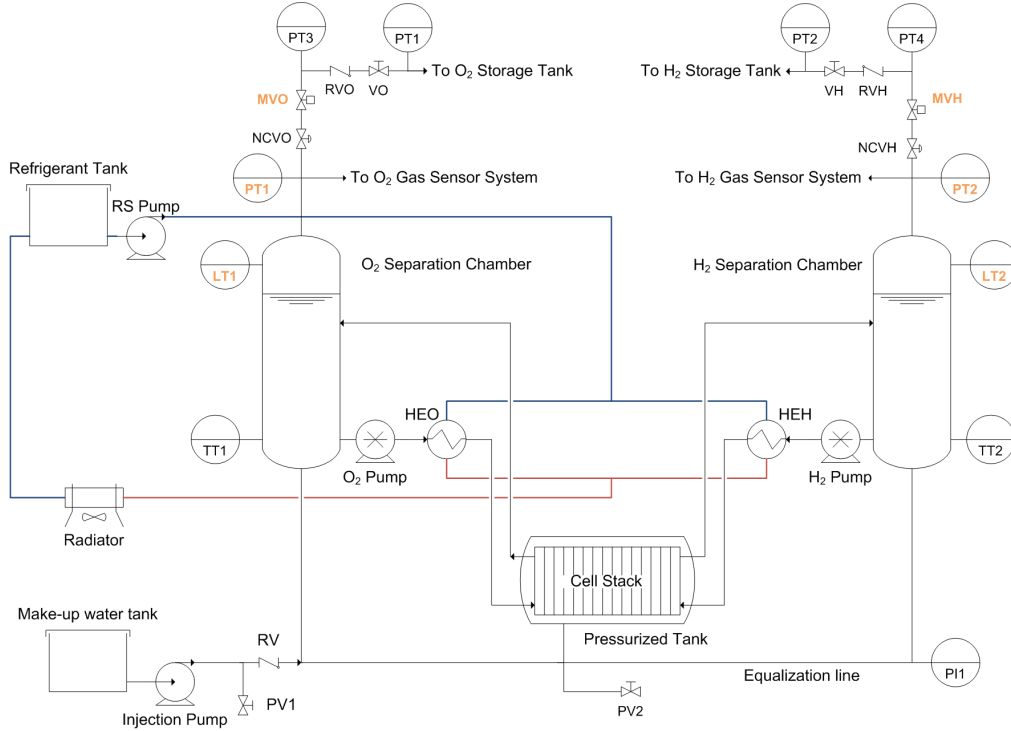


Figure 1: Piping and instrumentation diagram of the high-pressure alkaline electrolyzer. The main sensors and actuators explained in the text are highlighted in orange. Adapted from [19]

109 contamination process as [10] which is used into the phenomenological based
 110 semi-physical model reported in [19].

111 Electrolysis process happens in the electrolytic cell that is represented in
 112 Figure 2. Each cell is formed by two electrodes and a membrane which sep-
 113 arates both half cells. There are two driving forces for gas cross-permeation
 114 through this membrane. The first one is diffusion driven by differences in dis-
 115 solved gas concentration between the two half cells [24]. This phenomenon
 116 can be modelled on the basis of Fick's law as

$$\Phi_{c \rightarrow a, Fick} = D_{H_2} \frac{C_{H_2, c} - C_{H_2, a}}{z_{cell}}, \quad (1)$$

117 being $\Phi_{c \rightarrow a, Fick}$ the H_2 flux from cathode (c) to anode (a), D_{H_2} the diffusion
 118 coefficient of H_2 through the separator, $C_{H_2, x}$ the H_2 concentration in both
 119 half cells and z_{cell} the separator width. The presented equation corresponds

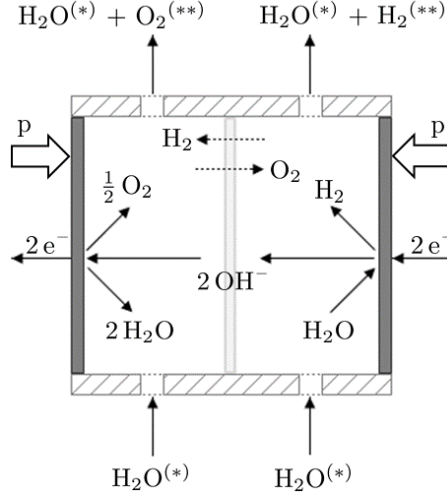


Figure 2: Scheme of the electrolytic cell with reactions. $H_2O^{(*)}$ represents KOH solution and $O_2^{(**)}$ and $H_2^{(**)}$ represent outputs that are contaminated with H_2 and O_2 , respectively. Taken from [19]

120 to the H_2 diffusion, a similar equation can be described for the O_2 .

121 The second cause of cross-contamination is the permeability of the elec-
 122 trolyte with dissolved gases due to differential pressure between both half
 123 cells. Based on Darcy's law, H_2 flux when cathodic pressure is higher than
 124 anodic one can be written as

$$\Phi_{c \rightarrow a, Darcy} = \epsilon_{H_2}^{Darcy} \frac{P_c - P_a}{z_{cell}}, \quad (2)$$

125 where $\Phi_{c \rightarrow a, Darcy}$ is the H_2 flux from cathode to anode when cathodic pressure
 126 P_c is greater than anodic pressure P_a . The H_2 permeability $\epsilon_{H_2}^{Darcy}$ depends
 127 on fluid properties and the concentration of dissolved H_2 . In case anodic
 128 pressure is greater than the cathodic one, a similar equation can be obtained
 129 for the O_2 contamination flux. Clearly, only one flux occurs at a time.

130 2.2. Control scheme

131 An alkaline electrolyzer requires several control loops for an efficient and
 132 safe operation. The control of both the refrigeration system and the make-up
 133 pump ensures a safe operation of the electrolyzer. Whereas, the H_2 produc-
 134 tion is controlled by the outlet valves. This paper is focused on the latter. A
 135 brief description of the other loops is described next.

136 The refrigeration system and the make-up pump are controlled indepen-
 137 dently by hysteresis cycles. These control loops, whose designs are not going
 138 to be treated in this paper, are defined by the following sets of constraints:

$$\begin{aligned} L_{\text{H}_2} \leq L_{\text{min}} \text{ and } L_{\text{O}_2} \leq L_{\text{min}} &\Rightarrow u_{\text{pump}} = 1, \\ L_{\text{H}_2} \geq L_{\text{max}} \text{ or } L_{\text{O}_2} \geq L_{\text{max}} &\Rightarrow u_{\text{pump}} = 0, \end{aligned} \quad (3)$$

$$\begin{aligned} T_{\text{H}_2} + T_{\text{O}_2} \geq 2 T_{\text{max}} &\Rightarrow u_{\text{RS}} = 1, \\ T_{\text{H}_2} + T_{\text{O}_2} \leq 2 T_{\text{min}} &\Rightarrow u_{\text{RS}} = 0, \end{aligned} \quad (4)$$

139 where L_{O_2} , L_{H_2} , T_{O_2} and T_{H_2} are the liquid solution levels and temperatures
 140 in O_2 and H_2 SCs, respectively. These variables are measured by the trans-
 141 mitters LT1, LT2, TT1 and TT2, respectively (see Figure 1). The limits im-
 142 posed are $L_{\text{min}} = 0.45$ m, $L_{\text{max}} = 0.5$ m, $T_{\text{min}} = 39.5$ °C and $T_{\text{max}} = 40.5$ °C.
 143 Finally, the control actions u_{pump} and u_{RS} manage the activation of the in-
 144 jection pump, the refrigeration system pump and the radiator, respectively.

145 Finally, energy management, with the consequent control of the current-
 146 voltage relationship, is intrinsically related to the power sources, so it is
 147 beyond the scope of this paper. Details on this topic can be found in [12, 17,
 148 25].

149 As previously indicated, in alkaline electrolysis, a pressure difference be-
 150 tween both half-cells generates the gas crossover. Therefore, the control ob-
 151 jective is to keep the liquid solution levels equalized in both SCs (measured
 152 by LT1 and LT2 in Figure 1) while H_2 and O_2 are delivered at a certain
 153 pressure (measured by PT1 and PT2 in Figure 1). This objective is achieved
 154 acting over two motorized outlet valves (MVO and MVH in Figure 1). The
 155 operating ranges for pressure p and electric current I are 0-7000 kPa and
 156 10-50 A, respectively. It is important to note that this electrolyzer, with an
 157 electrode area of $A_{\text{cell}} = 143$ cm², works in a current density j range between
 158 70-350 mA/cm² under the direct relationship

$$j = \frac{I}{A_{\text{cell}}}. \quad (5)$$

159 With the aim of having a suitable resolution in these wide operating ranges
 160 and considering the H_2 production capacity of 0.5 Nm³/h, needle-type outlet
 161 valves with a relatively small maximum flow coefficient, e.g., $C_v = 0.004$,
 162 must be used. In order to be able to control the system with only one valve
 163 per outlet line, the pressure in both storage tanks should be similar.

164 Another variable to be controlled is the difference between the liquid levels
 165 in both SCs, defined as

$$\Delta L = L_{\text{H}_2} - L_{\text{O}_2}. \quad (6)$$

166 This variable must be kept around a set-point $\Delta L_{\text{ref}} = 0$. This condition
 167 will contribute to the natural action of the equalization line circuit by keep-
 168 ing the pressure equalized on both sides of the membrane. In other words,
 169 if the control dynamics are slow enough, the equalization line ensures that
 170 the pressure in both SCs is almost the same, and the same happens in the
 171 electrolytic cells. As stated by Schalenbach et al [10], the ZirfonTM mem-
 172 brane is highly permeable to pressure differences, which was described in
 173 Section 2.1. These pressures P_c and P_a depend on the pressure of each SC
 174 and the pressure exerted by the column of liquid. In order to understand the
 175 effect of the liquid level difference in each SC, an example is presented next.
 176 A difference in level $\Delta L = 2$ mm represents a pressure difference of 25 Pa.
 177 Considering only this difference, a contaminating flow of H₂ from cathode
 178 to anode $\dot{n}_5 = 1.71 \times 10^{-9}$ kmol s⁻¹ occurs (see Figure A.11). The purity
 179 of the gases produced will depend on the rate of O₂ production. Therefore,
 180 with a low current density $j = 70$ mA/cm², an impurity of 0.24 % will be
 181 obtained. Finally, controlling the difference in level and pressure generates a
 182 high purity of the supplied gases. However, the absence of contamination is
 183 unreachable due to the natural diffusion that occurs in the studied process.

184 The control scheme proposed to achieve the objectives is presented in
 185 Figure 3. The controller produces two valve opening values, u_{H_2} and u_{O_2} ,
 186 taking values between 0 (minimum opening) and 10 (maximum opening).
 187 The control values are computed to ensure that

$$P_{\text{H}_2} \rightarrow P_{\text{ref}}, \quad (7a)$$

$$\Delta L \rightarrow 0. \quad (7b)$$

188 In normal operation, this pressure is set externally in order to follow smoothly
 189 the pressure of the storage tanks P_{tank} . Accordingly, the reference for the
 190 pressure P_{ref} is defined as

$$P_{\text{ref}} = P_{\text{tank}} + P_{\text{gap}}, \text{ subject to } |dP_{\text{ref}}/dt| < \alpha, \quad (8)$$

191 being α a rate limit in kPa/s. This rate limit ensures that a sudden change in
 192 the storage pressure does not generate an excessive variation in the pressure
 193 at both sides of the membrane, with the consequent cross-contamination.

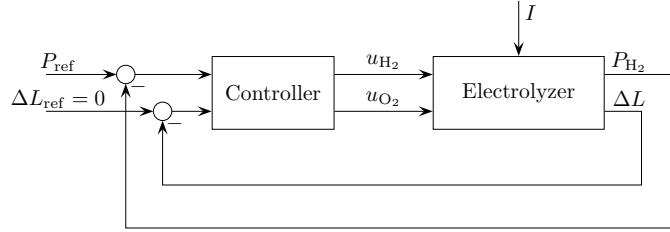


Figure 3: Proposed control scheme.

194 Under the assumption of similar pressures, P_{tank} is set equal to P_{H_2} . More-
 195 over, the pressure gap between P_{ref} and P_{tank} , $P_{\text{gap}} = 50$ kPa, is needed to
 196 compensate the action of the retention valves (RVO and RVH).

197 The described control loops act simultaneously and independently. For
 198 instance, when the make-up pump acts injecting water, the main loop devel-
 199 oped in this work equalizes the levels by either opening or closing the valves.
 200 Then, the action of the former (make-up pump) appears as a disturbance to
 201 the latter (outlet valves).

202 2.3. Reduced control-oriented model

203 A highly-detailed model for alkaline electrolyzers is given in [18, 19]. This
 204 model has 25 differential equations (i.e., 25 states) and 17 additional vari-
 205 ables, 50 structural parameters and 49 functional parameters. The basic
 206 structure of Ordinary Differential Equations (ODEs) is presented in the Ap-
 207 pendix and a complete description can be found in the aforementioned ref-
 208 erences.

209 Such a model is suitable for simulation purposes but not for control design.
 210 To this end, those variables that produce smaller effects on the controlled
 211 variables (ΔL and P_{H_2}) might be neglected under some assumptions and
 212 guaranteed conditions that are explained next.

- 213 • Although the ultimate goal is to maximize the purity of the gases,
 214 the concentrations of impurities are not taken into account for the
 215 controller, which is based on the liquid levels and the system pressure.
- 216 • In addition, despite having two paths of diffusion, i.e., through the
 217 membrane and through the equalization line, the latter is smaller than
 218 the former (10^6 times). This is mainly due to a longer path through the

219 equalization line (approximately 3 m) against just the thickness of the
220 membrane (approximately 5×10^{-4} m). Then, the diffusion through the
221 equalization line can be neglected along with the corresponding states.

222 • Moreover, under the hypothesis of reaching gas purities greater than
223 99%, saturation of pure gas in each cell can be assumed in order to
224 calculate diffusion across the membrane.

225 • Furthermore, according to the ideal gas law, the gas moles behave
226 equally no matter the substance, hence it only matters the accountancy
227 of the number of moles at each line.

228 • Finally, only the concentrations of pure gases in the electrolytic cells
229 and in the SCs can be considered.

230 Based on the previous assumptions, the model can be reduced to 14
231 states. The ODEs corresponding to these 14 states are listed in Table 1.
232 The rest of the states are considered constant while the parameters, which
233 are represented by algebraic equations, are not modified.

234 Different scenarios with pulse-type signals in the disturbances i and P_{tank}
235 and control inputs u_{H_2} and u_{O_2} were simulated to compare the responses
236 of the original model and the reduced COM. Figure 4 shows the results of
237 one of them when the initial operating conditions correspond to $I = 30$ A
238 and $P_{\text{H}_2} = 4000$ kPa (an operating point in the center of the considered
239 operating range) and only a pulse-type signal in the current is applied. The
240 duration of that pulse was 10 s and the amplitude was 30 A. In the top-plot
241 of Figure 4, the evolutions of the pressure P_{H_2} for the full original model
242 (solid black line) and the same pressure for the reduced COM (dashed red
243 line) can be observed. The second and third plots compare the evolutions of
244 the levels L_{H_2} and L_{O_2} , respectively. The bottom plot shows the difference of
245 levels ΔL for the original and the reduced models, respectively. The relative
246 errors¹ for the pressure and each level, and the absolute error, in case of the
247 level difference, can be seen in solid blue lines. In this last case, the nominal
248 values are close to zero and the relative error is impractical. Notice that the
249 maximum approximation error is 2×10^{-5} m in ΔL , which is quite small
250 comparing with the maximum value of this signal in Figure 4. In all cases,

¹The relative error is defined as $100 |y_{\text{original}} - y_{\text{reduced}}| / |y_{\text{original}}|$

Table 1: ODEs corresponding to the reduced nonlinear model.

State	Corresponding ODE
1	$\frac{d\bar{\rho}_3}{dt} = \frac{1}{V_{mix,I}} (\dot{n}_1 + \dot{n}_{22} + \dot{n}_6 - \dot{n}_3 - \dot{n}_5 - \dot{n}_{21} + r \sum_i \sigma_{i,1})$
2	$\frac{dx_{H_2,3}}{dt} = \frac{1}{N_I} \left(x_{H_2,1} \dot{n}_1 - x_{H_2,3} \dot{n}_3 - \dot{n}_5 + \dot{n}_6 + r_1 - x_{H_2,3} \dot{N}_I \right)$
3	$\frac{d\bar{\rho}_4}{dt} = \frac{1}{V_{mix,II}} (\dot{n}_2 - \dot{n}_{22} - \dot{n}_6 - \dot{n}_4 + \dot{n}_5 + \dot{n}_{21} + r \sum_i \sigma_{i,2})$
4	$\frac{dx_{O_2,4}}{dt} = \frac{1}{N_{II}} \left(x_{O_2,2} \dot{n}_2 + \dot{n}_5 - \dot{n}_6 - x_{O_2,4} \dot{n}_4 + \frac{r}{2} - x_{O_2,4} \dot{N}_{II} \right)$
5	$\frac{dN_{III}}{dt} = \dot{n}_3 + \dot{n}_7 - \dot{n}_9 - \dot{n}_{11}$
6	$\frac{dL_{Lg,III}}{dt} = \frac{1}{A_{SC}} \left(\dot{V}_3 + \dot{V}_7 - \dot{V}_9 - \dot{V}_{11} + \dot{V}_{b,III} \right)$
7	$\frac{dx_{H_2,III}}{dt} = \frac{1}{N_{III}} \left(x_{H_2,3} \dot{n}_3 + x_{H_2,7} \dot{n}_7 - \dot{n}_9 - x_{H_2,11} \dot{n}_{11} - x_{H_2,III} \dot{N}_{III} \right)$
8	$\frac{dN_{IV}}{dt} = \dot{n}_4 - \dot{n}_8 - \dot{n}_{10} - \dot{n}_{12}$
9	$\frac{dL_{Lg,IV}}{dt} = \frac{1}{A_{SC}} \left(\dot{V}_4 - \dot{V}_8 - \dot{V}_{10} - \dot{V}_{12} + \dot{V}_{b,IV} \right)$
10	$\frac{dx_{O_2,IV}}{dt} = \frac{1}{N_{IV}} \left(x_{O_2,4} \dot{n}_4 - x_{O_2,8} \dot{n}_8 - \dot{n}_{10} - x_{O_2,12} \dot{n}_{12} - x_{O_2,IV} \dot{N}_{IV} \right)$
11	$\frac{dP_{15}}{dt} = \frac{RT}{A_T L_{g,IX}} (\dot{n}_9 - \dot{n}_{15}) - \frac{P_{15}}{L_{g,IX}} \dot{L}_{g,IX}$
12	$\frac{dP_{16}}{dt} = \frac{RT}{A_T L_{g,X}} (\dot{n}_{10} - \dot{n}_{16}) - \frac{P_{16}}{L_{g,X}} \dot{L}_{g,X}$
13	$\frac{d\dot{n}_7}{dt} = \frac{1}{\tau_{equal}} (\dot{n}_{theo} - \dot{n}_7)$
14	$\frac{d\dot{n}_8}{dt} = \frac{1}{\tau_{equal}} (\dot{n}_{theo} - \dot{n}_8)$

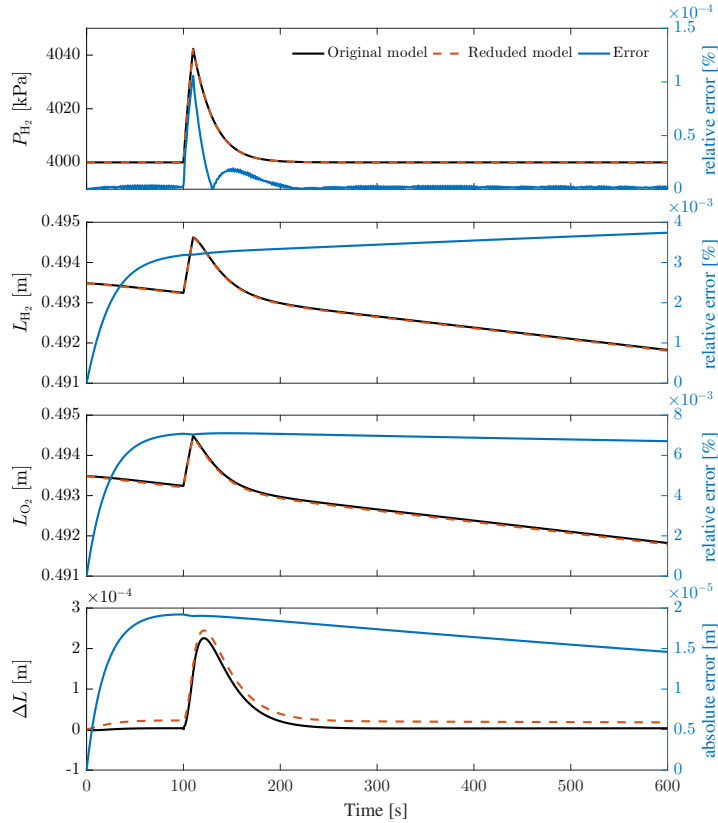


Figure 4: Comparison between the responses of the full nonlinear model and the reduced COM when a pulse of 10 s duration is applied in the current.

251 it can be observed that the pressure and levels in both separation chambers
 252 did not present differences while the difference in level has an increasing
 253 decoupling, although of small amount.

254 3. Control Design

255 In this section, two linear controllers are proposed for mitigating the cross-
 256 contamination of gases through the membrane in the alkaline electrolyzer
 257 presented in Section 2. The former is a classical PI control used frequently
 258 in industry, while the latter is a model-based \mathcal{H}_∞ optimal controller.

259 In both cases, a linear model of the electrolyzer is required, therefore
 260 the operating conditions of the electrolyzer must be defined. Assuming the

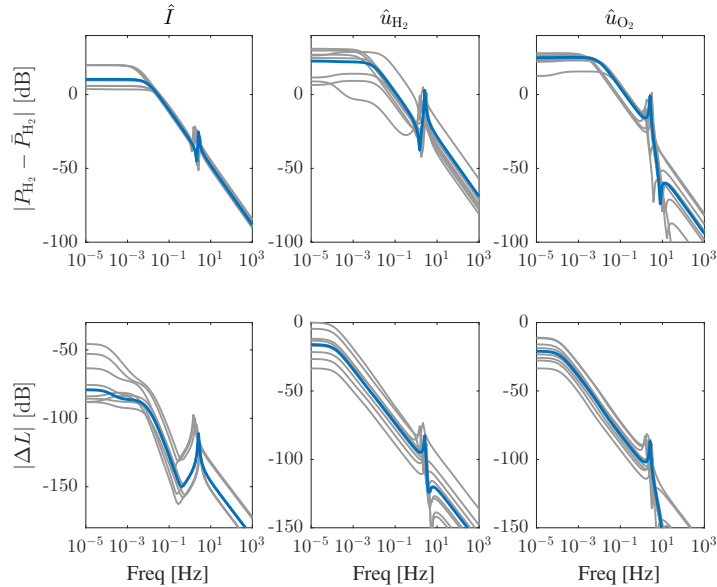


Figure 5: Frequency responses of the linearized model at several operating points (gray lines) and the nominal model $G(s)$ (blue lines).

261 control objective of tracking P_{ref} given in (8) and the regulation of ΔL around
 262 0 are satisfied, the operating conditions can be parameterized by the steady-
 263 state values of the tank pressure \bar{P}_{tank} and the current \bar{I} . Thus, the system
 264 operating region is defined as

$$\mathcal{O} = \{(\bar{P}_{\text{tank}}, \bar{I}) : 0 \text{ kPa} \leq \bar{P}_{\text{tank}} \leq 7000 \text{ kPa} \\ \text{and } 10 \text{ A} \leq \bar{I} \leq 50 \text{ A}\}.$$

265 Next, the reduced nonlinear model introduced in Section 2.3 is numeri-
 266 cally linearized at a representative operating point $(\bar{P}_{\text{tank}}, \bar{I}) \in \mathcal{O}$. To select
 267 this point, the linearization is performed over a grid of operating points in
 268 \mathcal{O} . The magnitude of the frequency responses for these operating points is
 269 shown in Figure 5 in gray lines and the selected nominal model is represented
 270 by a thicker blue line. This nominal model will be used to design the con-
 271 sidered linear controllers. It can be observed that there is no drastic changes
 272 in the frequency responses at different operating points. This fact suggests
 273 that linear controllers can achieve a suitable performance.

274 The selected nominal dynamics are approximated by the model

$$y(s) = G(s) \begin{bmatrix} \hat{I}(s) \\ \hat{u}(s) \end{bmatrix} = [G_d(s) \quad G_c(s)] \begin{bmatrix} \hat{I}(s) \\ \hat{u}(s) \end{bmatrix}, \quad (9)$$

275 where

$$\hat{u} = \begin{bmatrix} \hat{u}_{\text{H}_2} \\ \hat{u}_{\text{O}_2} \end{bmatrix} = \begin{bmatrix} u_{\text{H}_2} - \bar{u}_{\text{H}_2} \\ u_{\text{O}_2} - \bar{u}_{\text{O}_2} \end{bmatrix}, \quad y = \begin{bmatrix} P_{\text{H}_2} - \bar{P}_{\text{H}_2} \\ \Delta L \end{bmatrix}.$$

276 The variable \hat{u} is the vector of control inputs, and y is the vector of the
 277 controlled variables. The incremental current $\hat{I} = I - \bar{I}$ acts as a disturbance
 278 to be rejected. All of these variables are incremental values with respect
 279 to \bar{I} , \bar{u}_{H_2} , \bar{u}_{O_2} , and \bar{P}_{H_2} , where the last three variables are functions of the
 280 operating point $(\bar{P}_{\text{tank}}, \bar{I})$.

281 The first controller to be designed is based on classical PI design methods.

282 3.1. PI control

283 The system to be controlled, namely as $G_c(s)$, has two control inputs and
 284 two controlled outputs. It can be observed in Figure 5 that the control loops
 285 are coupled and a multivariable approach is required.

286 The simplest control approach consists in decoupling the loops and then
 287 designing two independent controllers [26]. For this purpose, the plant is
 288 right-multiplied by the inverse of its DC-gain, that is,

$$G_{\text{dec}}(s) = G_c(s)G_c(0)^{-1}. \quad (10)$$

289 Figure 6 compares the frequency response of the original and the decoupled
 290 plants, respectively. It can be observed that the diagonal elements dominate
 291 the dynamics and the off-diagonal present a small response in the frequency
 292 range of interest, as compared to the original nominal model $G_c(s)$.

293 The transfer functions corresponding to the diagonal elements of the de-
 294 coupled plant present a dominant dynamic behaviour similar to a first-order
 295 system, i.e., it can be approximated by

$$G_{\text{dec}}(s) \approx \begin{bmatrix} \frac{k_1}{s-a_1} & 0 \\ 0 & \frac{k_2}{s-a_2} \end{bmatrix}, \quad (11)$$

296 where $a_1 = -0.0576$ rad/s, $k_1 = 0.0576$, $a_2 = -7.817 \times 10^{-4}$ rad/s, and
 297 $k_2 = 7.817 \times 10^{-4}$. Consider the PI controller for each channel j ,

$$K_{\text{PI}}(s) = k_{p,j} \frac{s - b_j}{s}, \quad (12)$$

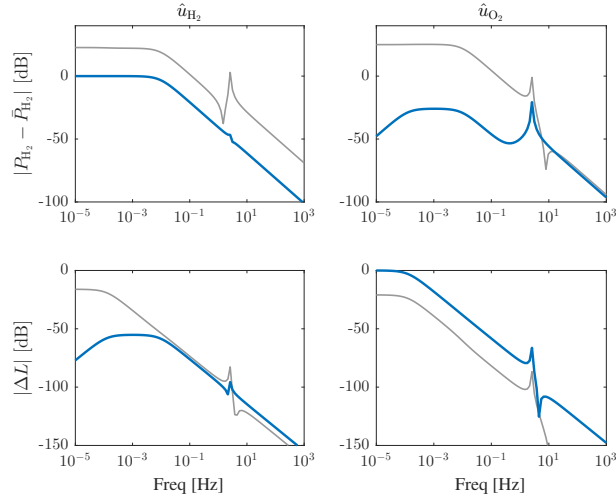


Figure 6: Frequency responses of the nominal plant $G_c(s)$ (gray lines) and decoupled plant $G_{\text{dec}}(s)$ (blue lines) used for the PI controller desing.

298 with $b_j = -k_{i,j}/k_{p,j}$ and being $k_{p,j}$ and $k_{i,j}$ the proportional and integral
 299 gains of the controller, respectively. Then, the controller parameters can be
 300 tuned by locating the zero b_j slightly at the left of the model dominant pole
 301 a_j and then adjusting the gain $k_{p,j}$ until a suitable closed-loop response is
 302 obtained.

303 The resulting closed-loop scheme combining the diagonal elements
 304 $G_{\text{dec},jj}(s)$, ($j = 1, 2$) and the PI controllers is stable for all values of $k_{p,j}$.
 305 Nevertheless, a limit on these parameters comes from the lack of perfect de-
 306 coupling, measurement noise levels, and the saturation of the control action.
 307 All these issues must be checked by simulation using the complete nonlinear
 308 model.

309 Next, a model-based robust controller will also be designed and compared
 310 with the previous one.

311 3.2. \mathcal{H}_∞ optimal control

312 Alternatively, the controller can be designed in the frame of multivariable
 313 optimal control. In this case, the control design objectives are expressed as

$$\min_{\tilde{K}(s)} \frac{\|z\|_2}{\|w\|_2}, \quad (13)$$

314 where z is a performance variable and w a disturbance. Therefore, the con-
 315 troller design consists in defining a control setup and in selecting z and w ac-
 316 cording to the control specifications with suitable weighting functions [27, 28].

317 In the electrolyzer case, tracking a pressure reference P_{ref} while rejecting
 318 the disturbance I is sought. Hence, the performance variable z represents
 319 the pressure and level errors, and the disturbance w , of the system pressure
 320 and the current, i.e.,

$$z = W_e(s)M(s) \begin{bmatrix} P_{\text{H}_2} - P_{\text{ref}} \\ \Delta L \end{bmatrix}, \quad w = W_u(s) \begin{bmatrix} P_{\text{ref}} \\ \hat{I} \end{bmatrix},$$

321 where

$$\begin{aligned} M(s) &= \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \frac{1}{s}, \\ W_e(s) &= \begin{bmatrix} k_{e,1} & 0 \\ 0 & k_{e,2} \end{bmatrix}, \\ W_u(s) &= \begin{bmatrix} k_{u,1} & 0 \\ 0 & k_{u,2} \end{bmatrix} \frac{s/0.1\omega_c + 1}{s/10\omega_c + 1}, \end{aligned}$$

322 being $k_{e,j}$, $k_{u,j}$ and ω_c design parameters. The weighting function $M(s)W_e(s)$
 323 penalizes the low frequencies of the pressure and level errors and $W_u(s)$ penal-
 324 izes the magnitude at high frequencies of the control actions. The closed-loop
 325 setup is shown in Figure 7.

326 The final controller is obtained after solving the optimization problem
 327 (13) and left-multiplying the resulting $\tilde{K}(s)$ by $M(s)$, that is,

$$K_\infty(s) = M(s)\tilde{K}(s). \quad (14)$$

328 This factorization is needed to ensure the existence of a stabilizing controller.

329 The order of the controller will be the order of the nominal model plus the
 330 order of all the weighting functions. Therefore, to simplify the real-time im-
 331 plementation, the order of $G(s)$ can be numerically reduced. As indicated in
 332 Section 3.1, the nominal model $G_c(s)$ exhibits a frequency response similar to
 333 a first-order system for each channel. Therefore, using a standard balanced-
 334 truncated reduction method [29], the linear time-invariant (LTI) nominal
 335 model of 14-th order is reduced by to a 2-nd order LTI model [27, 28]. The
 336 full and reduced models are compared in Figure 8. As observed in this figure,
 337 the reduced model is also dominated by two poles but is coupled unlike the
 338 model in Figure 6.

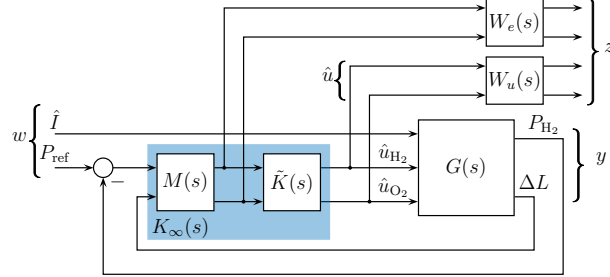


Figure 7: Control setup for the design of the \mathcal{H}_∞ controller.

339 4. Simulation results

340 Numerical simulations were performed with the previously designed con-
 341 trollers combined with the full-nonlinear model of the electrolyzer. The sim-
 342 ulations were performed in MatLab/Simulink with the variable-step solver
 343 Bogacki-Shampine (ode23), but other methods could also be used, e.g., Mul-
 344 tistage Adomian decomposition method [30, 31, 32]. Two different scenarios
 345 were considered and discussed below. In the first situation, a large depres-
 346 surization occurs while a constant electric current is applied. In the second
 347 scenario, the electrolyzer produces gases at constant pressure but the electric
 348 current fluctuates, as if it was provided by renewable energy sources. Previous
 349 reported results do not consider a dynamic model based on the phenomenol-
 350 ogy of the system for controller design, therefore a potential comparison with
 351 this work would be unfair.

352 The controllers were designed as indicated in Section 3. For the PI con-
 353 troller, the dominant poles of the decoupled plant are

$$a_1 = -0.0576 \text{ rad/s}, \quad a_2 = -0.00078 \text{ rad/s}.$$

354 Therefore, the controller zeros were located at $b_j = 1.05 a_j$ ($j = 1, 2$) resulting
 355 in the following parameters:

$$\begin{aligned} k_{i,1} &= 0.18, & k_{p,1} &= 3, \\ k_{i,2} &= 0.16, & k_{p,2} &= 200. \end{aligned}$$

356 For the \mathcal{H}_∞ controller, the design parameters in the weighting functions were
 357 set as

$$\begin{aligned} k_{e,1} &= 0.1, & k_{e,2} &= 4, \\ k_{u,1} &= 0.8, & k_{p,1} &= 0.8, \end{aligned}$$

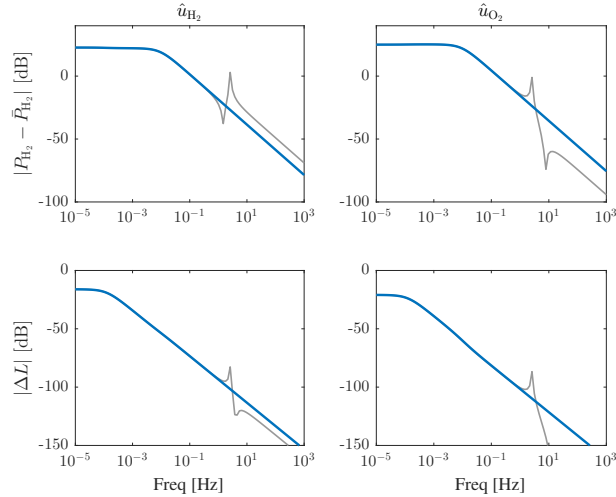


Figure 8: Frequency responses of the nominal plant $G_c(s)$ (gray lines) and of the reduced plant (blue lines) used in the \mathcal{H}_∞ controller design.

358 and $\omega_c = 0.7$ rad/s.

359 4.1. Scenario 1: Depressurization

360 This scenario analyzes a depressurization process caused by a sudden
 361 change in the tank pressure P_{tank} . This pressure drop can be caused by a
 362 preparation for a prolonged maintenance shutdown or by the system man-
 363 agement when low energy is forecasted.

364 Figure 9 shows the system responses with the PI controller (dashed lines)
 365 and the \mathcal{H}_∞ controller (solid lines). In the upper plot, a sudden change of
 366 P_{tank} from 7000 to 1000 kPa and the reference P_{ref} computed according to (8)
 367 with a rate limit of 5 kPa/s, can be observed. The current density is required
 368 to remain constant at $0.21\text{A}/\text{cm}^2$ (i.e., electric current $I = 30\text{A}$). Both con-
 369 trollers achieve a suitable pressure reference tracking. There are more visible
 370 differences between both controllers in the evolution of level difference ΔL .
 371 The \mathcal{H}_∞ controller achieves a faster convergence to the reference. On the
 372 other hand, impurity does not increase due to smooth control actions involv-
 373 ing equalized pressures on both sides of the membrane. Instead, the impurity
 374 decreases due to the production of gases at a lower pressure. The goal of this
 375 simulation is to achieve a depressurization without extra contamination dur-
 376 ing this process, which is reached with both controllers. In Figure 9, it can be
 377 seen that the control actions u_{H_2} and u_{O_2} do not exceed the actuator limits.

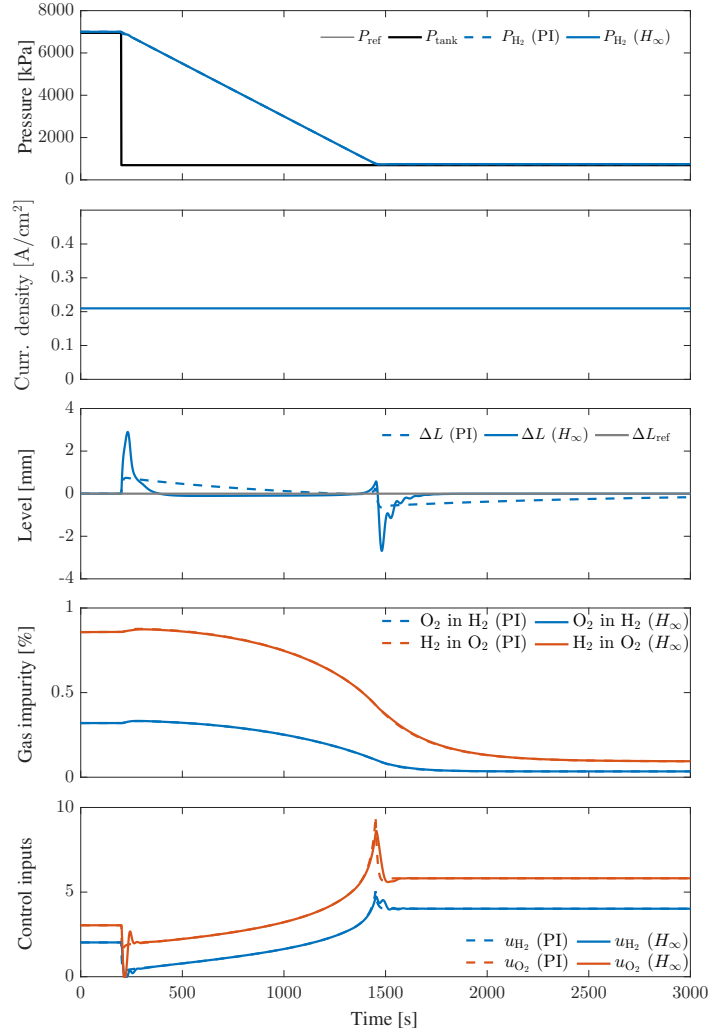


Figure 9: Simulation results corresponding to Scenario 1 using the PI controller (dashed lines) and the \mathcal{H}_∞ controller (solid lines).

378 4.2. Scenario 2: Electric current fluctuations

379 In this scenario, the current density changes while the pressure reference
 380 P_{ref} is kept constant. The simulations using both controllers are compared in
 381 Figure 10. As can be seen, valves openings virtually follow the fluctuation of
 382 the current density due to the direct relationship between current density and
 383 gas production. Both controllers manage to maintain the reference pressure

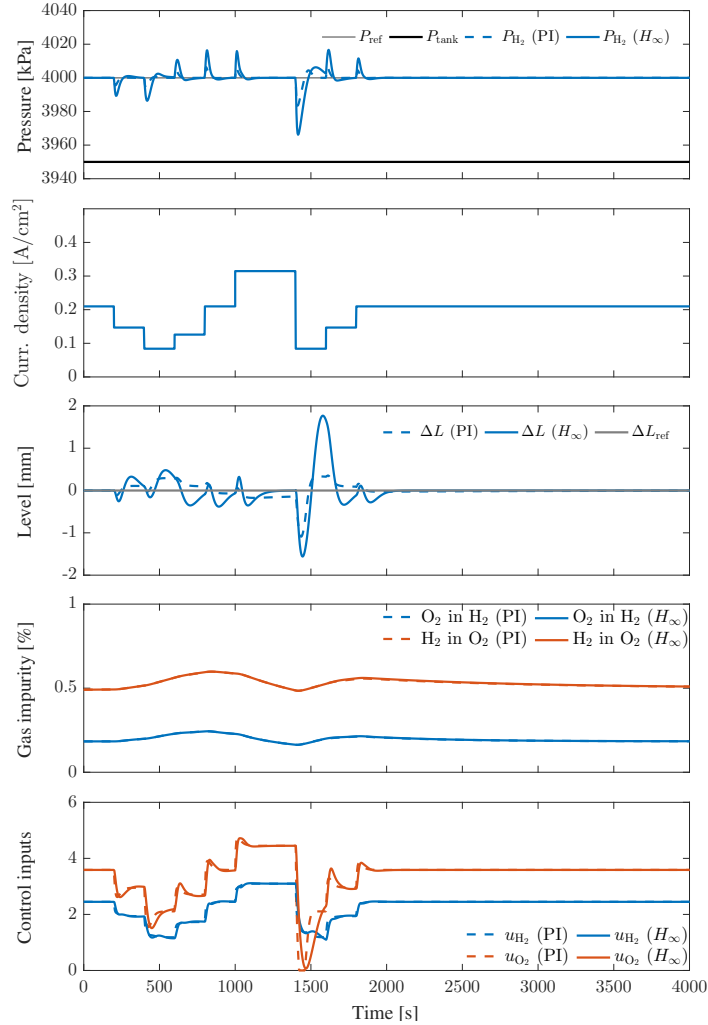


Figure 10: Simulation results corresponding to Scenario 2 using the PI controller (dashed lines) and the \mathcal{H}_∞ controller (solid lines).

384 with a maximum error of 0.5% and the level difference in less than 2 mm.
 385 Because of this, O_2 impurity, that is always the highest value, is below 1%.

386 4.3. Controller comparison

387 Particularly in scenario 1, the \mathcal{H}_∞ control has a higher transient error
 388 but converges to zero faster than in the PI case. Overall, the performance of
 389 both controllers is similar and depends on the tuning of the PI and the weight

390 selection for the \mathcal{H}_∞ control procedure. Both controllers were designed from
391 a common COM and seeking for the best performance/robustness compro-
392 mise. In case of the PI controller, the tuning procedure consists in adjusting
393 four parameters (the proportional and integral constants for each channel).
394 In the \mathcal{H}_∞ control, the design is based on an optimal algorithm and the
395 controller is tuned by the proper selection of a set of weighting transfer func-
396 tions. The PI controller might be preferred by some control engineers as it
397 is based on a more intuitive SISO tuning procedure. However, this method
398 relies on non-perfect decoupling that can affect the final closed-loop perfor-
399 mance. Instead, the \mathcal{H}_∞ controller requires more sophisticated design tools
400 but is designed directly from the MIMO model in an optimal way, based
401 on the performance/robustness weights that take care of low/high frequency
402 requirements.

403 5. Conclusions

404 In the quest to raise the operating pressure of alkaline electrolyzers, con-
405 trol strategies are needed to decrease gas cross-contamination and, conse-
406 quently, increase the purity of the supplied gases. In that sense, modelling
407 and control are key issues in operation and design improvements. This work
408 has proposed the design and comparison assessment of two different con-
409 trollers that were tested in closed loop with a high-fidelity nonlinear model
410 of the electrolyzer. They were able to maintain impurity below 1% in all
411 cases, keeping the liquid solution level difference between both separation
412 chambers below 4mm and a maximum pressure error of 0.5%.

413 Simulation results show that, with a suitable design, both controllers are
414 capable of achieving satisfactory performance. Design and implementation
415 issues will define which one is more practical. The design of the PI con-
416 troller requires less model information, but a diagonalization stage needs to
417 be made and the final parameters must be checked by extensive simulations.
418 The \mathcal{H}_∞ optimal controller algorithm is a multivariable system tool, the
419 design is systematic and only depends on the selection of adequate robust-
420 ness/performance weights.

421 6. Acknowledgement

422 This work has been partially funded by the Cheerful CSIC project (MHE-
423 200065).

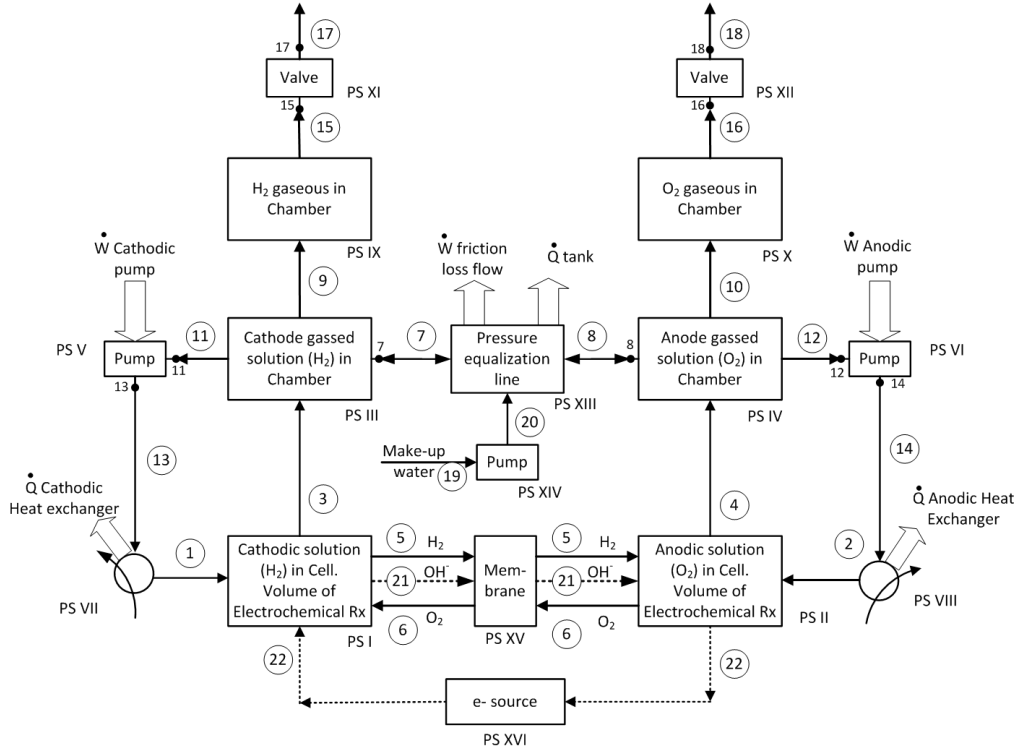


Figure A.11: Flow diagram with the PSs numbered in Roman. Mass flows are identified with numbers within circles. Taken from [19]

424 Appendix A. Summary of the complete nonlinear model

425 The PBSM is based on the flow diagram shown in Figure A.11, while a
 426 full description of the model is reported in [19]. A list of the ODEs governing
 427 the system behavior is given in Table A.2. This system has a clear symmetry
 428 between both lines: cathodic half-cell where H_2 is produced and anodic half-
 429 cell where O_2 is produced. Due to this symmetry, ODEs are similar between
 430 both lines. Therefore, in order to be concise, similar equations are presented
 431 once.

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Table A.2: Balance equations forming the model basic structure. Adapted from [19]

State	Corresponding ODE
1	$\frac{d\bar{\rho}_3}{dt} = \frac{1}{V_{mix,I}} (\dot{n}_1 + \dot{n}_6 - \dot{n}_3 - \dot{n}_5 + r_1 \sum_i \sigma_{i,1})$
2	$\frac{dx_{H_2,3}}{dt} = \frac{1}{N_I} \left(x_{H_2,1} \dot{n}_1 - x_{H_2,3} \dot{n}_3 - \dot{n}_5 + r_1 - x_{H_2,3} \dot{N}_I \right)$
3	$\frac{dx_{O_2,3}}{dt} = \frac{1}{N_I} \left(x_{O_2,1} \dot{n}_1 + \dot{n}_6 - x_{O_2,3} \dot{n}_3 - x_{O_2,3} \dot{N}_I \right)$
7	$\frac{dN_{III}}{dt} = \dot{n}_3 + \dot{n}_7 - \dot{n}_9 - \dot{n}_{11}$
8	$\frac{dL_{Lg,III}}{dt} = \frac{1}{A_{SC}} \left[\dot{V}_3 - \dot{V}_7 - \dot{V}_9 - \dot{V}_{11} + \dot{V}_{bubbles} \right]$
9	$\frac{dx_{H_2,III}}{dt} = \frac{1}{N_{III}} \left(x_{H_2,3} \dot{n}_3 + x_{H_2,7} \dot{n}_7 - \dot{n}_{H_2,9} - x_{H_2,11} \dot{n}_{11} - x_{H_2,III} \dot{N}_{III} \right)$
10	$\frac{dx_{O_2,III}}{dt} = \frac{1}{N_{III}} \left(x_{O_2,3} \dot{n}_3 + x_{O_2,7} \dot{n}_7 - \dot{n}_{O_2,9} - x_{O_2,11} \dot{n}_{11} - x_{O_2,III} \dot{N}_{III} \right)$
15	$\frac{dP_{15}}{dt} = \frac{RT}{A_T L_{g,IX}} (\dot{n}_9 - \dot{n}_{15}) - \frac{P_{15}}{L_{g,IX}} \dot{L}_{g,IX}$
16	$\frac{dx_{H_2,15}}{dt} = \frac{1}{N_{IX}} \left(x_{H_2,9} \dot{n}_9 - x_{H_2,15} \dot{n}_{15} - x_{H_2,15} \dot{N}_{IX} \right)$
17	$\frac{dx_{O_2,15}}{dt} = \frac{1}{N_{IX}} \left(x_{O_2,9} \dot{n}_9 - x_{O_2,15} \dot{n}_{15} - x_{O_2,15} \dot{N}_{IX} \right)$
21	$\frac{d\dot{n}_7}{dt} = \frac{1}{\tau_{equal}} (\dot{n}_{theo} - \dot{n}_7)$
23	$\frac{dN_{XIII}}{dt} = \dot{n}_{XIII,in} - \dot{n}_{XIII,out} + \dot{n}_{20}$
24	$\frac{dx_{H_2,XIII}}{dt} = \frac{1}{N_{XIII}} \left(x_{H_2,XIII,in} \dot{n}_{XIII,in} - x_{H_2,XIII,out} \dot{n}_{XIII,out} + A_{line} \Phi_{H_2} - x_{H_2,XIII} \dot{N}_{XIII} \right)$

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