processes

MDPI

Article

Modelling Acetification with Artificial Neural Networks and Comparison with Alternative Procedures

Jorge E. Jiménez-Hornero [1,*], Inés María Santos-Dueñas [2] and Isidoro García-García [2]

[1] Department of Electrical Engineering and Automatic Control, University of Cordoba, Campus Rabanales, 14071 Cordoba, Spain

[2] Department of Chemical Engineering, University of Cordoba, Campus Rabanales, 14071 Cordoba, Spain; ines.santos@uco.es (I.M.S.); isidoro.garcia@uco.es (I.G.)

* Correspondence: jjimenez@uco.es; Tel.: +34-957-212-079

Received: 28 May 2020; Accepted: 24 June 2020; Published: date

Supplementary material

Supplemental information to "Materials and Methods"

As explained in the main text, the process is conducted in a semi-continuous mode and each cycle is finished when the substrate (ethanol) is depleted to a preset extent. Then, the reactor is unloaded to an also preset volume and its residual content is used as inoculum in the next cycle, which is started by replenishing the tank with fresh medium, see Figure S1.

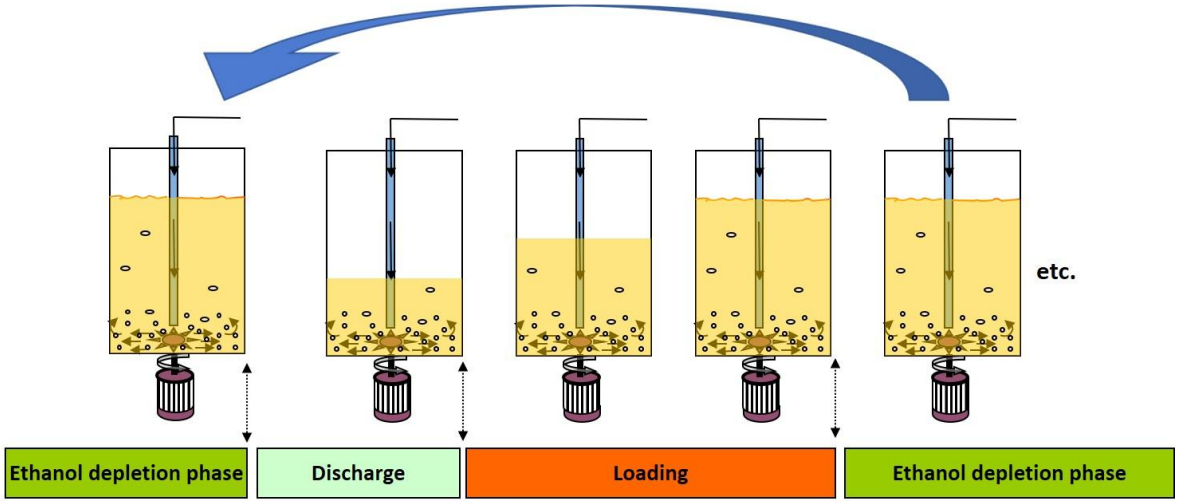

Figure S1. Typical semi-continuous working mode for vinegar production.

Normally, the operational variables used to control the process are as follows:

- The ethanol concentration in the raw material (E_i).
- The ethanol concentration at which the reactor is unloaded (E_{unload}).
- The volume of medium to be unloaded ($V_{unloaded}$).
- The mode of loading the bioreactor with fresh medium.

In practice, once the tank has been partially unloaded, a loading phase must be immediately started adding slowly fresh medium to avoid abrupt changes and exceedingly high local concentrations of ethanol [1,32]. In this regard, two different loading modes are used:

- Slowly refilling the tank to the final working volume is completed [24] (Figure S2). The mode referred as the "continuous" loading mode in Table 1.

- Slowly refilling the tank to the final working volume without exceeding a preset ethanol concentration [22] (Figure S3). The mode referred as the "semi-continuous" loading mode in Table 1.

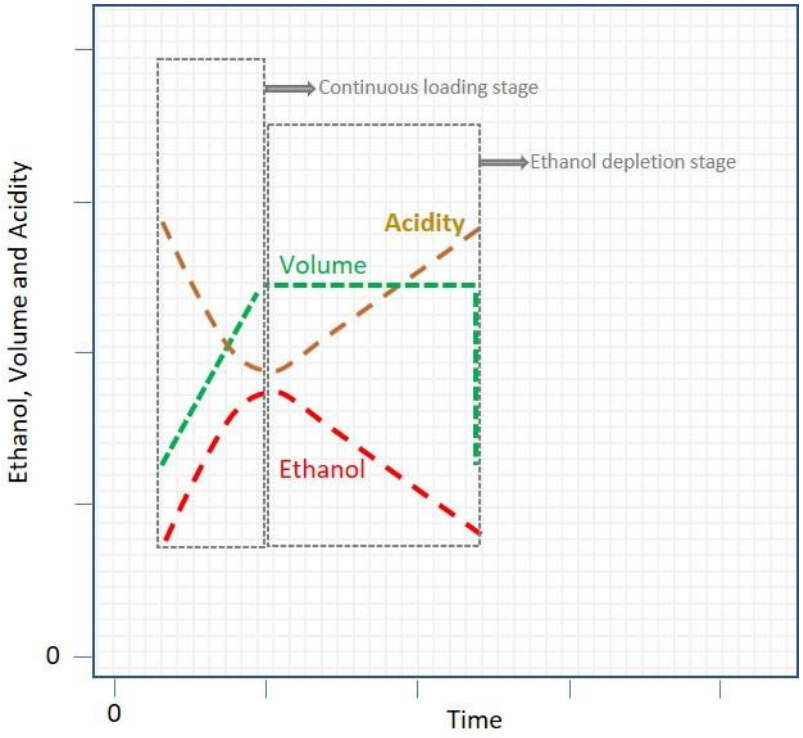

Figure S2. Continuous loading mode.

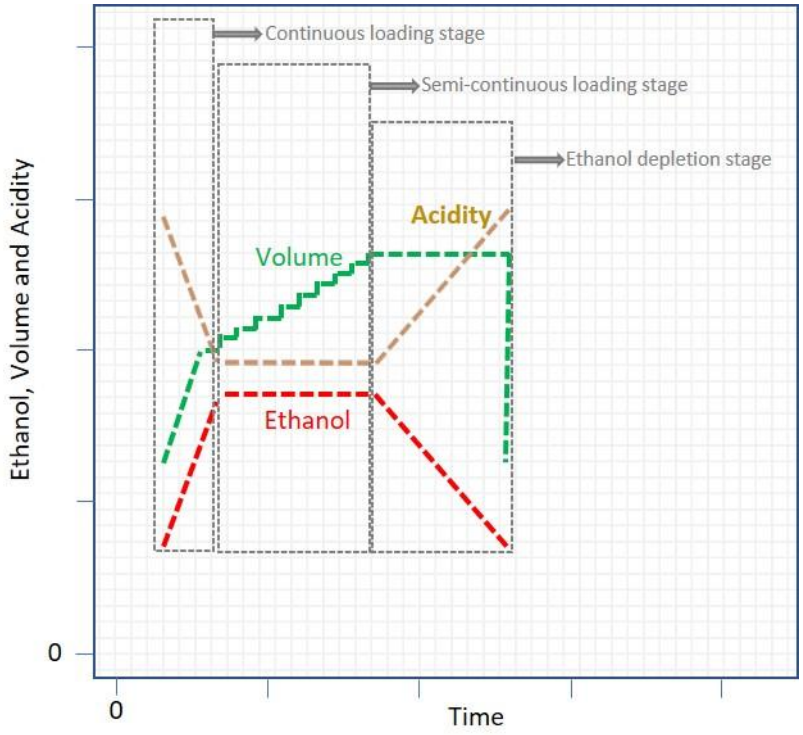

Figure S3. Semi-continuous loading mode without exceeding a preset ethanol concentration.

All the experiments have been carried out in a Frings 8 L fermentation tank at a constant temperature of 31 °C and an also constant air flow rate of 7.5 L air h^{-1} L^{-1} of medium. Once the desired

ethanol concentration for discharging was reached (E_{unload}), a percentage of the tank content is unloaded ($V_{unloaded}$). After that, a slowly loading phase is started following either the mode shown in Figure S2 or Figure S3, F_i being the feeding flow rate. A white wine (Montilla-Moriles type) containing 11.7 ± 0.3 % v/v ethanol (93 g ethanol L^{-1}) was used. The setup had a condenser to minimize volatile losses, so all the experiments were allowed to reach an acetic acid yield on ethanol of at least 95 % of the theoretical one [5,11].

By way of an example, next are shown some experimental results for one of the experiments with a continuous loading phase in Table 1, experiment 1 [5]:

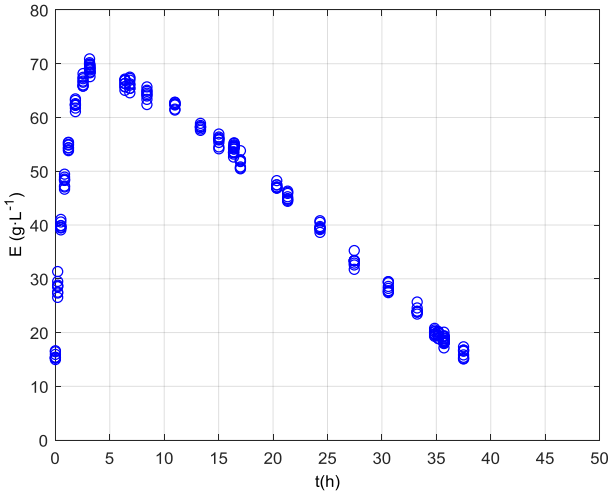

Figure S4. Ethanol concentration during cycle. Experiment 1, Table 1.

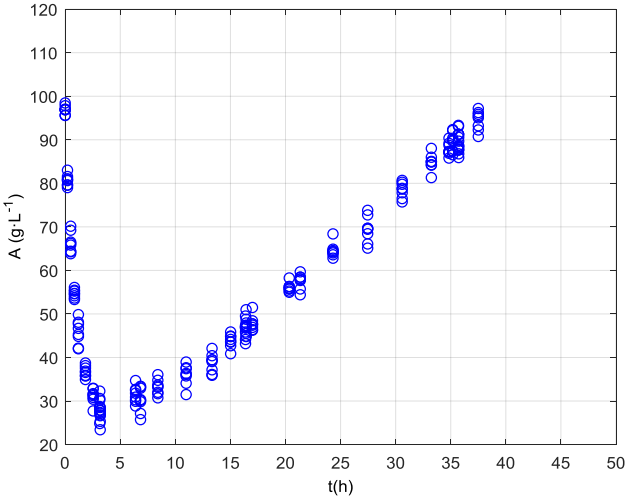

Figure S5. Acetic acid concentration during cycle. Experiment 1, Table 1.

Next, the results can represent some of the main aspects of the cycle:

- Mean ethanol consumption rate: 0.21±0.01 % v/v·h^{-1} (1.6±0.1 g EtOH·L^{-1}·h^{-1}).
- Acetic acid production rate: 15.1±0.5 g AcH·h^{-1}.
- Length of loading phase: 3.3±0.1 h.
- Total length of cycle: 37.5±1.1 h.
- Maximum ethanol concentration reached at the end of loading phase: 8.9±0.1 % v/v (69±0.8 g·L^{-1}).
- Maximum acetic acid concentration at the end of the cycle: 95±1 g·L^{-1}.

- At least ten cycles were repeated to obtain these results.

Similarly, also by way of example, next are shown some experimental results for another experiment in Table 1, in this case with the loading phase controlled by a maximum ethanol concentration, experiment 8 [5]:

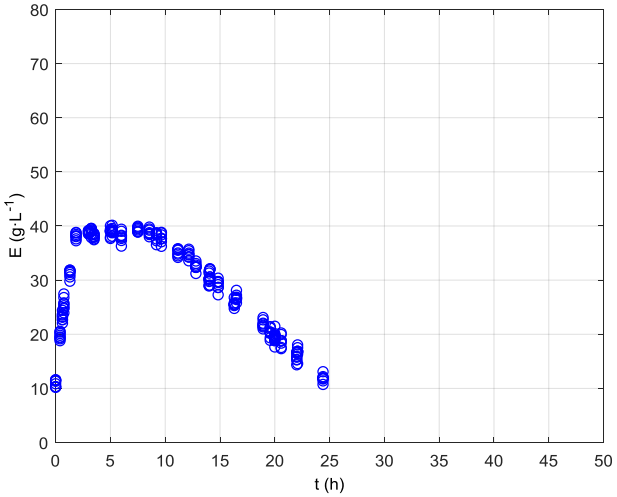

Figure S6. Ethanol concentration during cycle. Experiment 8, Table 1.

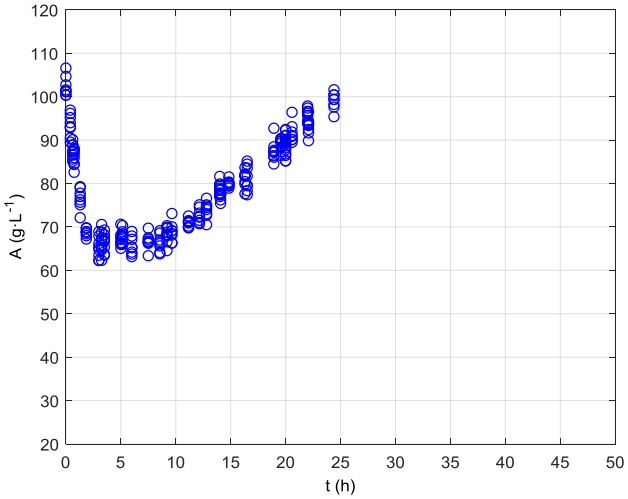

Figure S7. Acetic acid concentration during cycle. Experiment 8, Table 1.

Next, the results can represent some of the main aspects of the cycle:

- Mean ethanol consumption rate: 0.21±0.02 % v/v·h^{-1} (1.6±0.1 g EtOH·L^{-1}·h^{-1}).
- Acetic acid production rate: 14.8±0.5 g AcH·h^{-1}.
- Length of the continuous loading phase (first loading stage): 2.1±0.1 h.
- Length of the semi-continuous loading phase (second loading stage): 8.3±0.7 h.
- Total length of cycle: 27.3±0.4 h.
- Maximum ethanol concentration reached at the end of loading phase: 8.9±0.1 % v/v (69±0.8 g·L^{-1}).
- Maximum acetic acid concentration at the end of the cycle: 101±2 g·L^{-1}.
- At least ten cycles were repeated to obtain these results.

The issue of parameter identification is a difficult problem which has been discussed by authors elsewhere [8,9,32]; the last reference may offer a proper summary for it.

The polynomial black-box model was obtained using the experimental data shown in Table 2. In this case, all experiments used continuous loading strategy as drafted in Figure S3. A central composite design [11,24] was used; Table S1 shows the control factors and their levels and Table S2 the experimental plan.

Table S1. Control factors used in the Box–Behnken experimental plan and their levels [24].

Factor	*Code*	*Level*		
		(-1)	*(0)*	*(+1)*
Ethanol concentration at the end of the cycle, % v/v	E_{unload}	0.5	2	3.5
% unloaded volume	$V_{unloaded}$	25	50	75
Loading flow rate (L·min^{-1})	F_i	0.01	0.035	0.06

Table S2. Box–Behnken experimental plan and responses at different factor levels [11, 24]. E_{unload} (ethanol concentration at the end of the cycle, % v/v); $V_{unloaded}$ (% unloaded volume); F_i (loading flow rate (L·min^{-1})

Experiment	E_{unload}	$V_{unloaded}$	F_i	***Number of replications***
	Variable 1, X1	***Variable 2, X2***	***Variable 3, X3***	
1	3.5 (+1)	75 (+1)	0.06 (+1)	8
2	3.5 (+1)	75 (+1)	0.01 (-1)	10
3	3.5 (+1)	25 (-1)	0.06 (+1)	13
4	3.5 (+1)	25 (-1)	0.01 (-1)	21
5	0.5 (-1)	75 (+1)	0.06 (+1)	12
6	0.5 (-1)	75 (+1)	0.01 (-1)	8
7	0.5 (-1)	25 (-1)	0.06 (+1)	7
8	0.5 (-1)	25 (-1)	0.01 (-1)	7
9	3.5 (+1)	50 (0)	0.035 (0)	15
10	0.5 (-1)	50 (0)	0.035 (0)	10
11	2 (0)	75 (+1)	0.035 (0)	7
12	2 (0)	25 (-1)	0.035 (0)	16
13	2 (0)	50 (0)	0.06 (+1)	19
14	2 (0)	50 (0)	0.01 (-1)	11
15	2 (0)	50 (0)	0.035 (0)	12

This type of experimental design is visualized in Figure S8.

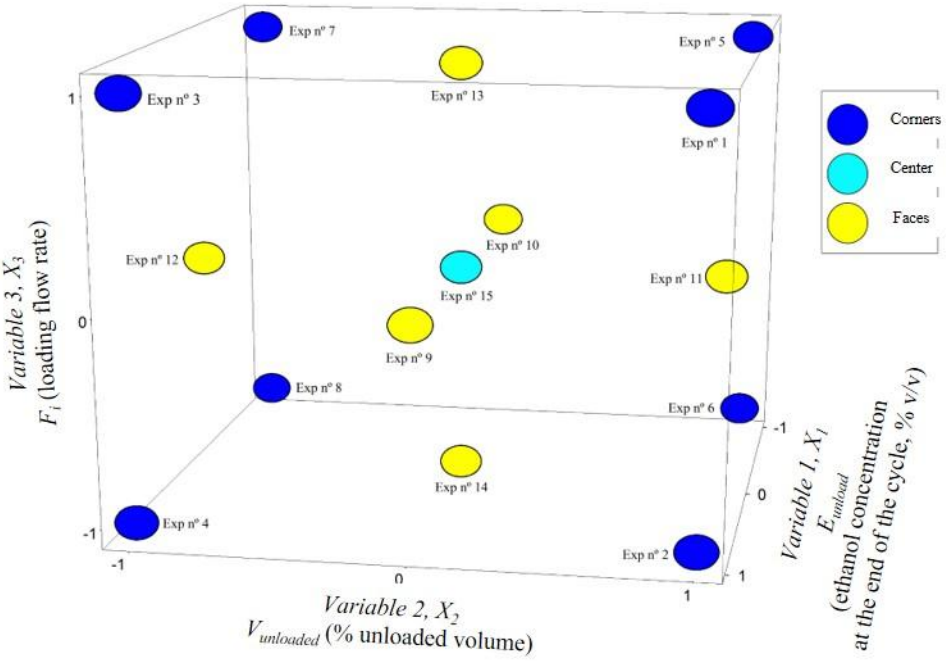

Figure S8. Central composite design used for polynomial regression.