Supplementary materials

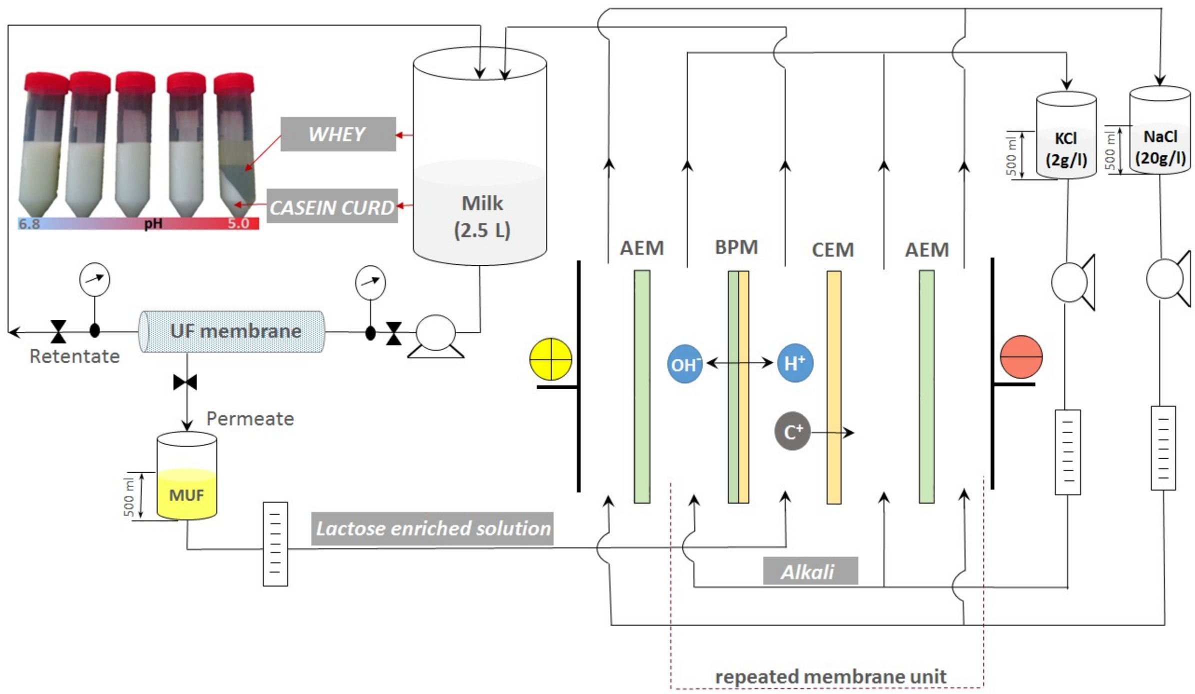

Fig.A1. Scheme of five-compartment bipolar membrane electrodialysis coupled with ultrafiltration. The final products are indicated inside the grey squares. C^{+} represents migrated cations.

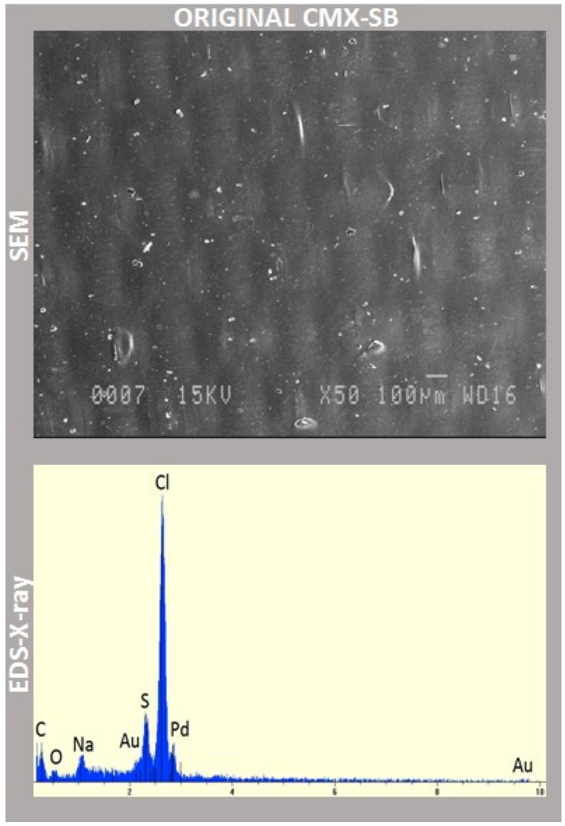

Fig.A2. Scanning Electron Microscopy (SEM) images and Energy Dispersive X-ray Spectroscopy images of original CMX-SB membrane.

Table A1. The quantities of references flows for EDBM-UF and acid/base processes of caseinate powder manufacturing scaled up to fulfill the functional unit, i.e. to produce 1000 kg eq. of caseinate powder.

	Incoming reference flows	Process*	Unit	Amount
Electromembrane treatment	Skim milk	A	kg	39098
	Deionized water	A, CW	kg	30529
	NaCl	A	kg	182
	Polysulfone	A	m^2	$6,07 \cdot 10^{-02}$
	Polyvinyl chloride	A	kg	$3,26 \cdot 10^{-02}$
	Styrene	A	kg	$7,51 \cdot 10^{-02}$
	o-Diethylbenzene	A	kg	$4,89 \cdot 10^{-03}$
	m-Diethylbenzene	A	kg	$4,89 \cdot 10^{-03}$
	Trimethylamine	A	kg	$3,89 \cdot 10^{-06}$
	Sulfuric acid	A	kg	$8,43 \cdot 10^{-06}$
	Anode	A	kg	$9,10 \cdot 10^{-05}$
	Polypropylene	A	kg	$9,10 \cdot 10^{-05}$
	Steel	A	kg	$9,10 \cdot 10^{-05}$

	Ultrafiltration module	A	items	$9{,}10\cdot10^{-05}$
	Electricity	A, CW, R, D	kWh	1245
	steam	R, D	kg	5882
Acid/Base treatment	Skim milk	A	kg	42821
	Deionized water	A, CW, R	kg	14753
	HCl (30 %)	A	kg	206
	NaOH (50 %)	R	kg	42
	Electricity	A, CW, R, D	kWh	370
	steam	R, D	kg	5882

*A stands for acidification process, CW for centrifugation/washing process, R for resolubilisation process and D for drying process.

Acidification process

The quantity of skim milk for the production of 1000 kg eq. caseinate powder were calculated taking into account the caseinate powder composition[19] and the purity of casein curd obtained after acidification process[12]. The quantity of deionized water consumed at acidification step for EDBM-UF treatment was calculated based on experimental data described in section 2.1, which includes water for the preparation of NaCl solutions of 2 g/L and 20 g/L concentrations. The deionized water consumed at the acidification step for acid/base treatment includes water for the preparation of 1.0 N HCl solution from 30 % solution. Quantity of NaCl needed for the preparation of solutions (2 g/L and 20 g/L) for EDBM module was calculated from the experimental data described in section 2.1. Polysulfone represents the basic material for the UF membrane and its quantity was calculated taking into account that the UF module includes 100 m^2 of membrane area, membrane lifetime is 3 years and there is 549557 kg of caseinate powder produced annually (5 days per week, overall 2080 hours). The polyvinylchloride, styrene and divinylbenzene (obtained by endothermic dehydrogenation of o-diethylbenzene and m-diethylbenzene[25]) represent the polymeric matrix of IEMs incorporated in EDBM module. These membranes are prepared by paste method and Mizutani et al. described the contribution of each above-mentioned component to the overall membrane polymeric matrix[26]. It was assumed that industrial EDBM module consists of 200 m^2 of CEM, 100 m^2 of AEM and 100 m^2 of BPM having a lifetime of one year. The weights of dry membranes were measured and were taking into account during calculations of incoming fluxes related to the IEMs. The trimethylamine and sulfuric acid represent the components of ion-exchange groups[26], which are fixed on the polymeric matrix of IEMs. Their quantities were calculated based on the membrane lifetime, dry weight and ion-exchange capacity. The anode, polypropylene (material of intermembrane gaskets) and steel (material of cathode) represents the materials of EDBM module, having a lifetime of 20 years as well as UF module. The electricity flux at the acidification step for EDBM-UF treatment consists of the power consumed by the EDBM module (858 kWh) and power consumed by pumps of EDBM (1.36 kWh) and UF modules (16.58 kWh). The electricity consumed during conventional acidification comprises only mixing tank power consumption (2 kW) considering an acidification time of 10 min. The quantity of HCl was determined concerning 1.60 mol of acid per kg. of casein obtained after acidification[12].

Centrifugation-Washing process

The deionized water consumed for the washing of casein curd after acidification process represents 25 % of the incoming acidified milk[27]. The electricity consumption was calculated based on the power consumption of the centrifuge-decanter (55 kW) and washing tank (1.5 kW) considering the four centrifugation-decantation steps of 10 min each and three washing steps of 20 min each[28].

Resolubilisation process

The deionized water consumed during resolubilisation step for the acid/base treatment includes water for the preparation of 1.0 N NaOH solution from 50 % solution. The quantity of NaOH were determined based on the assumption of 0.5 mol of NaOH per kg. of casein solids[19]. The electricity consumption for both treatments comprises the consumptions related to the grinding operations of casein curd on the colloid mill (27 kW) during 20 min and dissolution of casein curd in NaOH solution in the dissolution tank (2 kW) during 45 min[28]. The steam consumption for heating of the sodium caseinate solution (up to 65 °C) to reduce its viscosity was calculated based on specific heat (4.0 kJ/kg°C), temperature difference (45 °C), specific enthalpy of steam evaporation (2108.1 kJ/kg) and mass of sodium caseinate solution.

Drying process

The electricity and steam consumptions for the both treatments were calculated from Hui et al.[29] considering that the moisture of the caseinate powder is 3.8 %[19]. The air consumption is not taken into account due to its recycling during powder production.

Geographical sensitivity test of electricity supply mix

Results of Figure 6 are contextualized in the Canadian province of Quebec, where 95.3 % of electricity production is from hydropower, which makes this province a leader in terms of sustainability of electricity production in comparison to other provinces and countries[20]. On Figure 6 electricity (included in "others") accounts for an insignificant share of environmental impacts when comparing acid-base and EDBM-UF processes. However, how will the eco-profile change in different geographical locations with different electricity production mix? Considering that EDBM-UF process requires about three times more electricity (1245 kWh) than acid/base process (370 kWh) for the production of 1000 kg eq. of caseinate powder, are the conclusions still maintained in a different geographical context?

To answer these questions, a sensitivity analysis was performed taking into account different geographical localization of the electricity mix supply as an inventory flow. The results of sensitivity test demonstrate that sustainable leader of the caseinate production is Quebec using hydroelectricity as an energy source having the lowest emissions as compared to other considered regions (Fig.A3). Moreover, EDBM-UF process releases 8.9 units of % less of CO_2 eq. as compared to acid/base process. One could notice that countries of Eastern Europe have very similar environmental impacts with difference between the both studied scenarios is 8.7 units of %. In this region, electricity is mainly produced from natural gas, hydropower and nuclear power having relatively low emissions. For instance, in Russian Federation, 50.1 % of electricity is produced from natural gas, 17.1 % from hydropower, 16.3 % from nuclear power and just 15.2 % and 0.1 % from coal and oil respectively[17]. Similar impacts (8.6 units of % difference between both scenarios) were demonstrated for countries of South America using almost the same sources of energy as in Eastern Europe. The emissions associated with caseinate manufacturing in Western Europe are slightly higher as compared to above regions, which leads to decrease of differences between EDBM-UF and acid/base processes to 8.3 units of %. This could be related to the fact that electricity production in Western Europe is quite diversified. For instance, in France more than 70 % of electricity comes from nuclear power while in Norway it mainly comes from hydropower (96.1 %)[17]. More than 40 % of German electricity is derived from coal as in Greece and Denmark, which is associated with significant environmental issues[17]. However, these issues are addressed to the development of the electricity production from renewable sources. Indeed, there is relatively wide implementation of renewable energy sources in certain countries of Western Europe such as Denmark (46 %), Germany (21 %), Iceland (29 %), Italy (21 %), Spain (26 %), etc[17]. Thus, the diversification of electricity production in Western Europe with the aim of environmental burden decrease could explain the relatively low impacts of electromembrane process for the caseinate production. The use of non-renewable energy sources in North America, Oceania, Africa and Persian Gulf countries (mainly coal and natural gas) for the electricity production leads to the higher environmental burden during caseinate manufacturing. In these regions, the differences between EDBM-UF and acid/base technologies are about 7.5 units of %. The least favorable regions for caseinate manufacturing from a sustainable point of view are China and Asia. For instance, in China with the largest electricity production industry (5719 TWh/year)[45] the main energy source is coal (> 70 %). Consequently, the differences between both studied scenarios of protein manufacturing are just about 6 units of %.

The similar trends could be observed for the ecosystem (6.9 – 8.9 units of %) and resources (4.6 – 8.9 units of %) impact categories between EDBM-UF and acid/base processes at different geographical locations (Fig.A.4). Though the general trends for the human health (5.3 – 8.0 units %) are similar to above categories, there is one particular region where the EDBM-UF process has 9.0 units of % higher impacts than acid/base one (Fig.A4). This region is Asia (without China) where the electricity production is associated with the important impacts on human respiration due to the emissions of inorganic pollutants from coal power production.

Thus, the sensitivity test revealed that the novel electromembrane process of caseinate production has less environmental burden compared to conventional process using chemicals no matter the electricity supply mix.

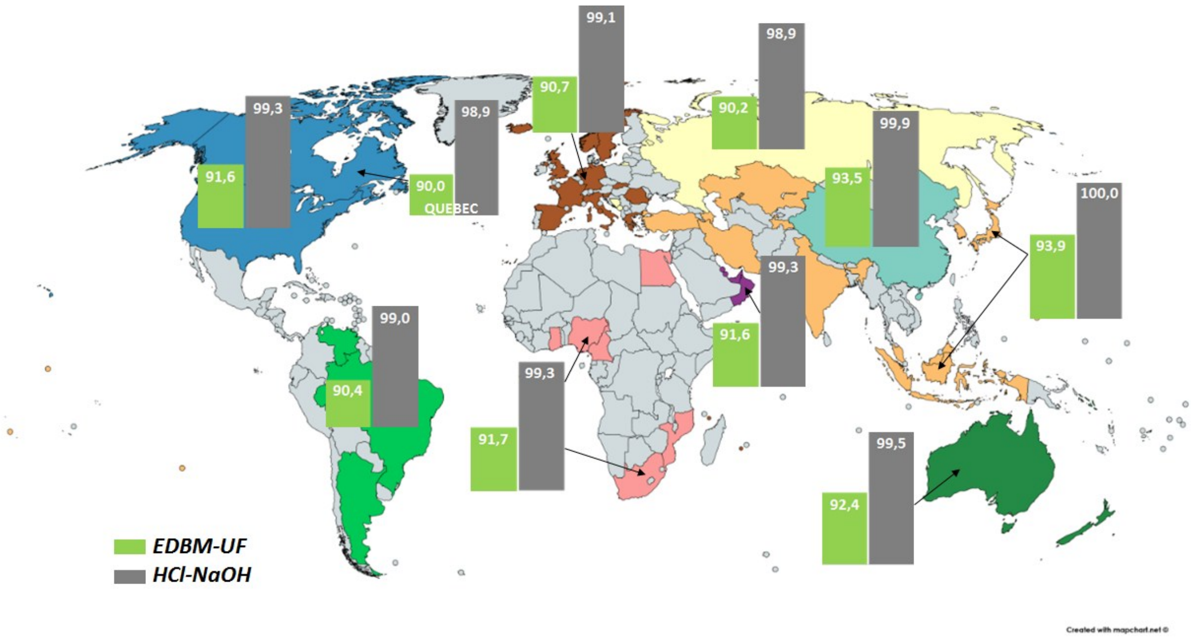

Fig.A3. Relative contributions (in %) of caseinate powder manufacturing by electromembrane and acid/base processes to the climate change (IPCC 2013) concerning the geographical localization of electricity inventory flow.

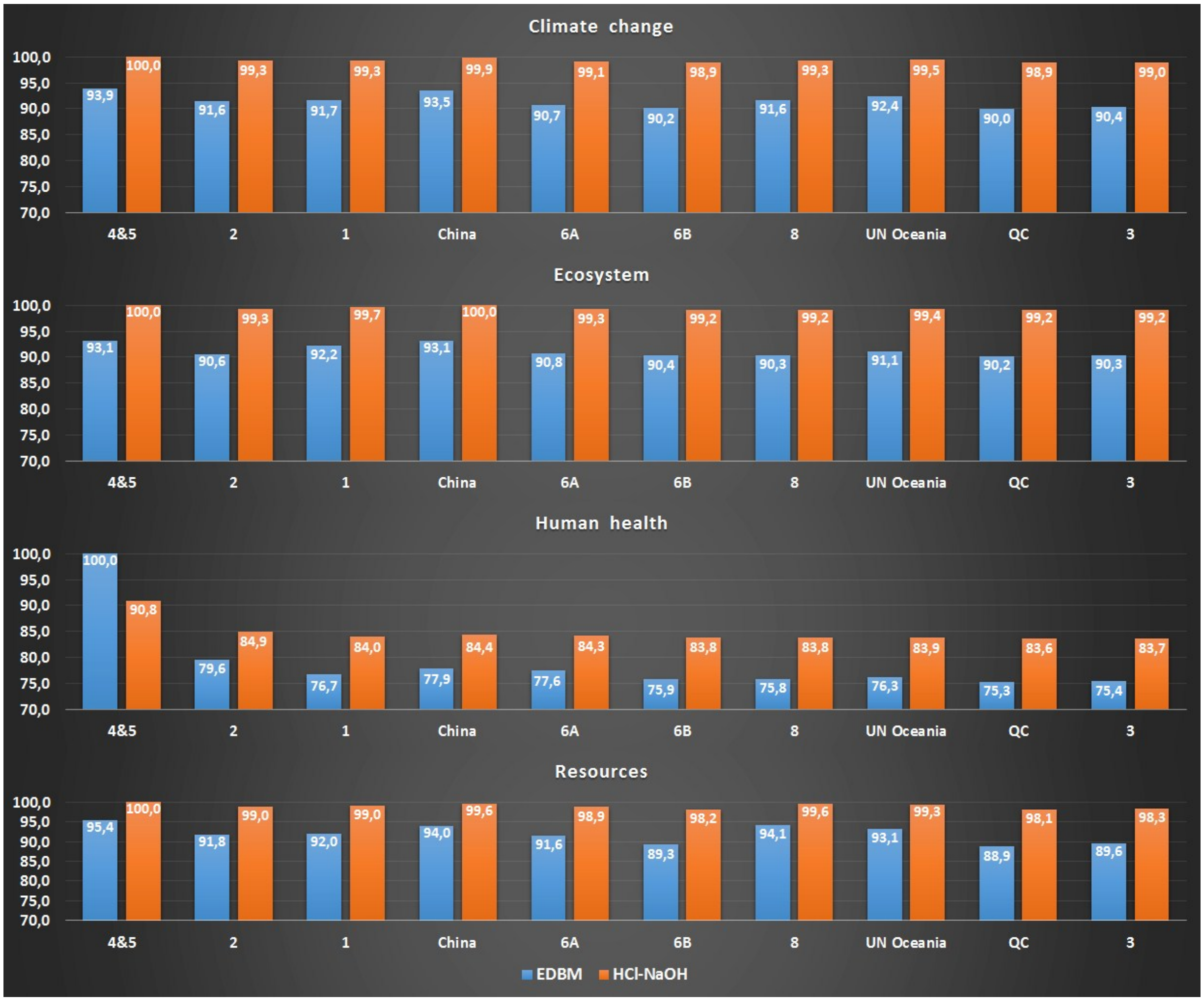

Fig.A4. Relative contributions (in %) of caseinate powder manufacturing by electromembrane and acid/base processes to the impact categories: climate change (IPCC 2013), human health (Impact 2002+), ecosystem quality (Impact 2002+) and resources (Impact 2002+) at different geographical locations of electricity production. Definition of areas: 4&5 – Azerbaijan, India, Indonesia, Iran, Japan, Kazakhstan, Malaysia, Tajikistan, Turkey; 2 – Canada (without Quebec), USA; 1 - Cameroon, Egypt, Ghana, Mozambique, Nigeria, South Africa; 6A - France, Germany, Greece, Iceland, Italy, Netherlands, Norway, Spain, Sweden, United Kingdom, Romania, Slovakia and Slovenia; 6B - Russia, Montenegro, Bosnia and Herzegovina; 8 - Bahrain, Oman, Qatar, UAE; UN Oceania – Australia, New Zealand; 3 – Argentina, Brazil, Venezuela.