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Investigation of the anaerobic digestion of cosmetic industrial wastes: Feasibility and perspectives / Fiore, S.; Demichelis, F.; Chiappero, M.; Onofrio, M.. - In: JOURNAL OF ENVIRONMENTAL MANAGEMENT. - ISSN 0301-4797. - ELETTRONICO. - 299:(2021), p. 113678. [10.1016/j.jenvman.2021.113678]

Availability:

This version is available at: 11583/2926032 since: 2021-09-21T14:51:10Z

Publisher:

Elsevier

Published

DOI:10.1016/j.jenvman.2021.113678

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<http://dx.doi.org/10.1016/j.jenvman.2021.113678>

(Article begins on next page)

1 **INVESTIGATION OF THE ANAEROBIC DIGESTION OF COSMETIC**
2 **INDUSTRIAL WASTES: FEASIBILITY AND PERSPECTIVES**

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11 **Abstract**

12 This study assessed the anaerobic digestion (AD) of wastes deriving from cosmetics production:
13 sludge from onsite wastewater treatment plant (sWWTP), residues of shampoo/conditioner
14 (RSC) and sludge from mascara production (MS), considered as single substrates and as
15 mixture according to the produced amounts (54 %-wt sWWTP, 31 %-wt RSC, 13 %-wt MS,
16 plus 2 %-wt food waste from the canteen, FW). Total COD (COD_T) was 624-1436 g O₂/kg vs,
17 while soluble COD was 5-23 %-wt of COD_T. AD tests at 35 °C achieved the following biogas
18 yields: 0.10 Nm³/kg_{vs} (70 %-v/v methane) for sWWTP; 0.07 Nm³/kg_{vs} (62 %-v/v methane) for
19 RSC; 0.04 Nm³/kg_{vs} (67 %-v/v methane) for MS. The mixed substrates underwent physico-
20 chemical pre-treatments (thermo-alkaline, TA: 120 min at 50 °C; thermo-alkaline-sonication,
21 TAS: 15 min at 40 kHz and 80 °C, both based on the addition of 0.08 g NaOH per each g of
22 total solid in the substrate), reaching 64-66 % disintegration rate, and AD tests (5 %-wt dry
23 substance) at 35 and 52 °C. Biogas yields were (for TA and TAS respectively): 0.22 and 0.20
24 Nm³/kg_{vs} (62-70 % methane); 0.21 and 0.19 Nm³/kg_{vs} (66-66 % methane) at 52 °C. At both
25 temperatures, methane yields considerably improved (+71-100 %), compared to mixed

26 untreated substrates, and 5-8 %-wt total solids reductions were observed. A technical-economic
27 scale-up assessment completed the research. The energy analysis highlighted the crucial role of
28 TA pre-treatment in achieving the process energetic sustainability. The economic analysis
29 showed that the AD of the considered cosmetic waste could be sustainable anyway, thanks to
30 the savings related to the disposal of the digestate compared to current waste management costs.

31 **Keywords:** biogas, cosmetic waste, economic analysis, pre-treatment, scale-up.

32 **1. Introduction**

33 According to the European Parliament regulation No. 1223/2009 (European Parliament, 2009),
34 cosmetic products may include “any substance or mixture intended to be placed in contact with
35 the external parts of the human body with the aim of cleaning, perfuming, protecting, keeping
36 them in good conditions”. The same EU regulation bans the use of hazardous substances in
37 cosmetic products to ensure the highest protection of human health. Growing interest for the
38 sustainability of raw materials employed to manufacture cosmetic products (Bom et al., 2019)
39 has been involving in recent years the consumers, cosmetics industries and academia.
40 Nonetheless the major concern on ensuring the highest safety and sustainability of cosmetic
41 products, the European Waste Catalogue (EWC) and the list of wastes in art. 7 of Directive
42 2008/98/CE often define hazardous and mark with an asterisk the EWC codes assigned to
43 wastes deriving from the manufacturing of fats, grease, soaps, detergents, disinfectants and
44 cosmetics (a sub-chapter of wastes deriving from organic chemistry processes). Liquid and solid
45 wastes originating from cosmetic products manufacturing are usually incinerated or landfilled,
46 while wastewater is typically treated in onsite treatment plants (Demichelis et al., 2018a),
47 because of the high concentrations of chemical oxygen demand (COD), suspended solids, fats,
48 oils and detergents (Abidemi et al., 2018). Enhanced onsite wastewater treatment operations are
49 necessary in cosmetic industries to mitigate the impact of the treated effluent on the receiving

50 water bodies (De Melo et al., 2013). The presence of scarcely biodegradable compounds in
51 cosmetics wastewater lead to the adoption of different treatment strategies, as:

52 1. *the enhancement of soluble COD to promote subsequent biological processes* through
53 physico-chemical pre-treatments (Demichelis et al., 2018a), catalytic wet peroxide oxidation
54 (Bautista et al., 2010) or combined light/Fe⁰/H₂O₂ processes (Muszyński et al., 2019);

55 2. *the decrease of total COD* through flocculation/coagulation (Kim et al., 2020), electro-
56 coagulation (Müller Tones et al., 2020), combined electro-coagulation and heterogenous photo-
57 catalysis (Boroski et al., 2009) or advanced oxidation processes, as electro-chemical oxidation
58 (Awad and Ghany, 2013), Fenton oxidation (Bautista et al., 2007), photo Fenton-like treatment
59 and heterogenous photo-catalysis (De Andrade et al., 2020);

60 3. *the application of biological processes highly efficient towards the removal of high organic*
61 *loads*, as membrane bioreactors (MBR) (Banerjee et al., 2016; Friha et al., 2014; Monsalvo et
62 al., 2014), upflow anaerobic sludge blanket (UASB) (Puyol et al., 2011) and biofilm (Fan and
63 Wang, 2017) reactors.

64 On the grounds of the performed literature survey on cosmetic industrial waste management,
65 two issues appeared noteworthy: state-of-the-art literature exclusively focused on cosmetic
66 wastewater treatment, leaving out any other waste; cosmetic wastewater treatment applied at
67 full-scale only involve, to our knowledge, physico-chemical pre-treatments preliminary to
68 active sludge processes, or MBR and UASB designs, while all other options, even if
69 characterized by high COD removal performances, are not yet mature at full-scale and
70 technically and economically feasible (Zhen et al., 2017). The aim of this work is to fill the gap
71 investigating the feasibility of the AD of cosmetic waste. Compared to existing literature, this
72 research has the two general elements of novelty: it is specifically focused on cosmetic waste
73 and not on cosmetic wastewater; it explores the feasibility of the AD of unconventional scarcely
74 biodegradable substrates. In details, this work investigated the treatment of the cosmetic wastes

75 (303 t/y) produced by L'Oréal Group in a plant located in Italy; specifically, sludge from the
76 onsite wastewater treatment plant (sWWTP), residues of shampoo and conditioner (RSC,
77 hazardous), sludge from mascara production (MS, hazardous) and food waste (FW) from the
78 canteen, considered singularly and as mixture according to produced amounts. At the moment
79 of this research, these waste materials were landfilled or incinerated. This work is the follow-
80 up of a previous study (Demichelis et al., 2018a), which investigated the technical feasibility of
81 different physico-chemical pre-treatments, based on chemicals (NaOH or HCl), heat (50-90 °C)
82 and sonication, and their various combinations for different times and temperatures, on the same
83 wastes. The performances of the pre-treatments were compared for the single wastes according
84 to the disintegration rate (DR, i.e., the % enhancement of soluble COD) (Li et al., 2012) and the
85 best results were obtained for thermo-alkaline (TA) treatment at 50 °C for 120 min and thermo-
86 alkaline-sonication (TAS) at 80 °C for 15 min, reaching DR equal to 64 % for TA and 66 % for
87 TAS. Further AD tests at 35 °C on the waste mixture, feeding 5 %-wt dry substance after TA
88 and TAS, showed 44-48 % increase of methane production (0.13 Nm³/kg_{VS}) and 5-7 %-wt
89 reduction of waste amounts compared to the untreated wastes. The study was concluded by a
90 preliminary analysis of the scale-up at full plant scale of TA and TAS, reaching the economic
91 feasibility (14-20 % savings compared to the adopted waste management operations), with TA
92 exhibiting lower investment and operational costs compared to TAS. Related to the described
93 previous study (Demichelis et al., 2018), the common aspects (apart of the considered cosmetic
94 wastes) with the present work are just the adoption of TA and TAS as pre-treatments and the
95 investigation of AD at 35 °C feeding 5 %-wt dry substance; the follow up presented in this work
96 has five significant improvements: 1. the characterization of the samples was enhanced; 2. the
97 investigation of AD explored mesophilic (35 °C) and thermophilic (52 °C) conditions; 3. AD
98 tests were performed on single wastes at 35 °C and on their pre-treated (TA and TAS) mixture
99 at 35 °C and at 52 °C (while the previous study considered AD tests only of the waste mixture

100 at 35 °C); 4. the performances and kinetics of the AD processes were analyzed through the
101 modified Gompertz model; 5. finally, the overall process (pre-treatment + AD) was scaled-up
102 based on energy and economic analyses, to explore the economic sustainability of the value
103 chain.

104 **2. Materials and methods**

105 *2.1 Substrates characterization*

106 The wastes analyzed in the research were sampled in 2016 at L'Oréal Saipo Industriale (L'Oréal
107 Group) plant located near Turin (Italy), producing shampoo and conditioner, mascara, and
108 make-up powders for the mass market in 34 countries. Waste production in 2015 was equal to
109 303 t and included: sludge from onsite biological wastewater treatment plant (sWWTP) (EWC
110 070612), residues of shampoo and conditioner production (RSC) (EWC 070608*), sludge from
111 mascara production (MS) (EWC 070601*) and food waste from the canteen (FW) (EWC
112 200108). At the moment of the research, sWWTP was landfilled, RSC and MS were incinerated,
113 and FW was sent to composting. In this work, waste samples were considered individually and
114 as mixture (in the following defined "Mix") according to the relative amounts produced in the
115 plant in 2015 (54 %-wt sWWTP, 31 %-wt RSC, 13 %-wt MS, 2 %-wt FW).

116 The characterization involved 5 samplings for each waste, along 9 months, to assess any
117 variability in composition. The measured parameters were: total solids (TS) and volatile solids
118 (VS) according to reference procedures (APHA-AWWA-WEF, 2012); pH through a pH 340
119 WTM pH-meter; total chemical oxygen demand (COD) and soluble chemical oxygen demand
120 (sCOD) according to a reference method (Raposo et al., 2008); ammonium through Nanocolor
121 Ammonium Test 1-05 reagents and a UNICAM Helios Alpha UV-vis spectrophotometer;
122 elemental analysis was performed by means of a CHNSO Thermo Fisher Flash 2000 Analyzer
123 EA 1112. Unfortunately, only one replicate was possible in the elemental analysis of RSC and
124 MS. The digestate was analyzed at the end of the AD tests measuring TS and VS and the lower

125 heating value (LHV) according to UNI EN 15170:2009 method, adopting benzoic acid as
126 reference material. LHV analyses involved 5 replicates for each sample.

127

128 *2.2. Pre-treatments*

129 The pre-treatments tests involved the mixed wastes (Mix) according to the procedures
130 optimized in a previous work (Demichelis et al., 2018a). Specifically, in both cases the NaOH
131 was added to the Mix (0.08 g/g of TS of the substrate), and the two pre-treatments were
132 performed as follows. The thermo-alkaline (TA) treatment at 50 °C for 120 min in a Julabo
133 Corio-C thermostatic bath and termo-alkaline-sonication (TAS) at 40 kHz in a VWR USC
134 300TH device at 80 °C for 15 min. Full details about the optimization of the pre-treatments and
135 the results achieved on single waste materials are available in the cited study (Demichelis et al.,
136 2018a).

137

138 *2.3. Anaerobic digestion tests*

139 AD tests were carried out at 35 °C on the single waste materials (FW excluded) and the Mix,
140 and at 52 °C on the Mix. In all cases in batch mode, feeding 5 %-wt TS and adopting substrate-
141 inoculum ratio equal to 1:1 based on VS. The inoculum was provided by two local facilities: a
142 WWTP performing mesophilic AD of the sludge and a plant performing thermophilic AD of
143 the organic fraction of municipal solid waste; before the tests, the inoculum was stored at 20 °C
144 and degassed for 7 days. The mesophilic inoculum was characterized by 1.71 %-wt TS, 75 %
145 VS/TS and pH 7.3; the thermophilic inoculum by 2.65 %-wt TS, 60 % VS/TS and pH 6.9. The
146 AD tests were performed in 0.5 L Pyrex Duran glass bottles (0.4 L working volume), connected
147 by 6 mm Teflon tubes to 2.5 L gas-tight Tedlar bags. Each reactor was purged with nitrogen for
148 1 min to remove air from the headspace. A 55 L Julabo-Corio-C thermostatic water-bath
149 allowed to control the temperature of the digesters. For each substrate (the single wastes and

150 the Mix), 12 reactors were engaged: 2 blanks (containing only the inoculum), 3 for biogas
151 measurement and 3 for methane measurement. Unfortunately, only one replicate was available
152 for methane measurement at 52 °C. During the tests, each reactor was manually shaken three
153 times per day for 5 days per week. Biogas and methane volumes were measured 5 days per
154 week through water displacement (10 g/L NaOH aqueous solution was employed to wash CO₂
155 from the reactors used for methane measurement) and referred to standard temperature and
156 pressure (0 °C, 1 atm). The AD tests were declared completed when marginal biogas production
157 was below 1 %.

158

159 *2.4. Performance and kinetics analysis*

160 The performance and kinetics of AD were analyzed according to Gompertz modified model
161 (Nguyen et al., 2016) according to (eq. 1).

$$162 \quad B(t) = P \exp \left\{ - \exp \left[\frac{R_{max} e}{P} (\lambda - t) + 1 \right] \right\} \quad (1)$$

163 where $B(t)$ is the cumulative methane production (Nm³ kgVS⁻¹) at time t (day), P is the methane
164 potential of the substrate (Nm³ kgVS⁻¹), R_{max} is the maximum methane production rate (Nm³
165 kgVS⁻¹ d⁻¹), λ is the lag phase (day), e is the base of the natural logarithm. The kinetic parameters
166 were estimated using non-linear least square regression analysis by means of the SOLVER tool
167 pack of Microsoft Excel.

168

169 *2.5. Scale-up assessment: reactor design and energy balance*

170 The scale-up of the process applied on the Mix at 35 °C in a continuously fed digester was
171 based on two scenarios: AD without any pre-treatment (S1) and after TA pre-treatment (S2),
172 considering the amount of waste produced at the moment of the research (303 t/y) (section 2.1).
173 Two sub-scenarios, based on the amount of available waste, were also considered for S2: a.
174 investigating the size of the digester necessary to process 303 t/y of waste mixture; b.

175 investigating the minimum size of the digester necessary to produce enough methane to achieve
 176 energy self-sufficiency (thermal and electric energy). The reactor design parameters were
 177 derived from the results of the pre-treatments and of the AD tests presented in section 3.3.
 178 The energy analysis was performed to estimate the net thermal energy (TE_{net}) and the net
 179 electrical energy (EE_{net}), expressed in MJ/year and considering one year of operations, as in eq.
 180 1 and 2, adopting the parameters and calculations detailed in Table 1.

181

$$182 \quad TE_{net} = TE_{CHP} - TE_{pre} - TE_{AD} - TE_{loss} \quad (1)$$

$$183 \quad EE_{net} = EE_{CHP} - EE_{mix} \quad (2)$$

184

185 Where TE_{CHP} [MJ/year] is the thermal energy obtained from the biogas in a combined
 186 heat&power (CHP) unit; TE_{pre} [MJ/year] is the thermal energy spent during the TA pre-
 187 treatment; TE_{AD} [MJ/year] is the thermal energy required to heat the substrate prior to AD;
 188 TE_{loss} [MJ/year] is the energy lost through the walls of the reactor. EE_{CHP} [MJ/year] is the
 189 electricity obtained from the biogas in the CHP unit, and EE_{mix} [MJ/year] is the electrical energy
 190 needed for the mixing.

191

192 **Table 1.** Parameters and calculations adopted in the energy analysis

| Parameter | Calculations | Notes | Reference |
|---|--|--|----------------------------|
| net thermal energy (TE_{net}) [MJ/year] | $TE_{net} = TE_{CHP} - TE_{pre} - TE_{feed} - TE_{loss}$ | | |
| heating pre-treatment (TE_{pre}) | $TE_{pre} = m_{mix} C_p (T_{pre} - T_{amb}) t \cdot 365 / \eta$ | specific heat capacity (C_p): 4.18 MJ/(ton·K); efficiency of the heat exchange (η): 0.65 pre-treatment temperature (T_{pre}): 50 °C ambient temperature (T_{amb}): 15 °C; treatment time (t): 2 hours/day | Gomez-Camacho et al., 2021 |
| heating feeding AD (TE_{AD}) | $TE_{AD} = m_{mix,5\%} C_p (T_{AD} - T_{amb}) 365 / \eta$ | AD temperature (T_{AD}): 35 °C; ambient temperature (T_{amb}): 15 °C | Gomez-Camacho et al., 2021 |
| thermal loss AD (TE_{loss}) | $TE_{loss} = U_{wall} A_{reactor} (T_{AD} - T_{amb}) \cdot 31.536$ | natural heat convection h air: 10 W/(m ² ·K); | Gomez-Camacho et al., 2021 |

| | | | |
|--|---|---|----------------------------|
| | | concrete: conductivity (λ) 2.3 W/(m·K); thickness (s): 0.25 m; insulation: λ : 0.037 W/(m·K); s: 0.08 m, U_{wall} : 0.422 W/(m ² ·K); surface reactor (A_{reactor}): 89 m ² LHV methane (LHV _{CH4}): 35.8 MJ/m ³ ; CHP thermal energy efficiency (η_{TH}): 0.4 methane production (Q_{CH4}) [m ³ /day] | |
| thermal energy from CHP (TE _{CHP}) | $TE_{\text{CHP}} = Q_{\text{CH4}} \eta_{\text{TH}} \text{LHV}_{\text{CH4}}$ 365 | | Metcalf&Eddy, 2013 |
| net electrical energy (EE _{net}) [MJ/year] | $EE_{\text{net}} = EE_{\text{CHP}} - EE_{\text{mix}}$ | | |
| electricity mixing AD (EE _{mix}) | $EE_{\text{mix}} = P_{\text{mix}} t V_{\text{reactor}}$ 31536 | power input (P_{mix}): 0.02 kW/m ³ ; reactor volume (V_{reactor}): 64 m ³ mixing time (t): 15/60 hours | Gomez-Camacho et al., 2021 |
| electricity from CHP (EE _{CHP}) | $EE_{\text{CHP}} = Q_{\text{CH4}} \eta_{\text{EL}} \text{LHV}_{\text{CH4}}$ 365 | CHP electrical efficiency (η_{EL}): 0.4 | Metcalf&Eddy, 2013 |

193

194 2.6. Scale-up assessment: economic analysis

195 A preliminary economic assessment was performed according to (Demichelis et al., 2018a and
196 b; Chiappero et al., 2019) to estimate the profitability of the two scenarios detailed in section
197 2.5. The profitability assessment was based on the following parameters: return of investment
198 (ROI), i.e., the ratio between the annual net profit and the initial investment; net present values
199 (NPV), and payback time (PBT). A plant lifetime equal to 20 years and a 5 % discount rate on
200 the future cash flows were considered. The economic assessment involved capital costs,
201 operational costs, and revenues. The annual net profit was determined as the difference between
202 the revenues and the operational costs; the amortization of the initial capital cost was not
203 considered in the present analysis. The capital costs included the fixed capital costs (FCC), and
204 the working capital costs (WCC) assumed equal to 6.5% of FCC [4]. The cost of land was not
205 considered since the installation was placed inside the cosmetic industrial plant. The operational
206 costs were composed by cost of the NaOH for the pre-treatment and of the water for the dilution
207 of the Mix, by thermal and electrical energy (obtained from the energy assessment), and labor
208 costs. The revenues derived from the surplus electricity from biogas co-combustion in the CHP

209 unit, and from the saved costs related to the disposal of the digestate (0.12 €/kg) compared to
 210 the costs of the current waste disposal for sWWTP (0.15 €/kg), RSC and MS (0.31 €/kg), and
 211 FW (0.11 €/kg) (Demichelis et al., 2018a).

212 3. Results and discussion

213 3.1. Substrates characterization

214 The results of the characterization of the studied cosmetic wastes (Table 2) showed that all
 215 substrates could be eventually considered suitable for AD based on their physico-chemical
 216 features (VS above 75 %, TS below 35 %, pH not too far from neutrality), even if the values of
 217 C/N were rather lower (respectively 10.71 for sWWTP, 5.49 for RS, 5.87 for MS and 14.40 for
 218 FW) than the values (20-35) usually recommended for AD (Metcalf&Eddy, 2013). Moreover,
 219 the low values of COD_s/COD_T witnessed the need of pre-treatments aimed at increasing the
 220 amount of soluble organic substances. sWWTP showed the highest variability in COD values,
 221 which was not unexpected; in fact, the onsite WWTP was fed by wastewater deriving from the
 222 washing of reactors and pipes in the productive lines of L'Oréal plant, where industrial
 223 production was organized in batches of different cosmetic products.

224 The Mix (54 %-wt sWWTP, 31 %-wt RSC, 13 %-wt MS, 2 %-wt FW) was characterized by
 225 14.06 % TS, 83.68 % VS/TS and C/N equal to 8.86.

226 **Table 2.** Characterization of the cosmetic wastes (*only one replicate available)

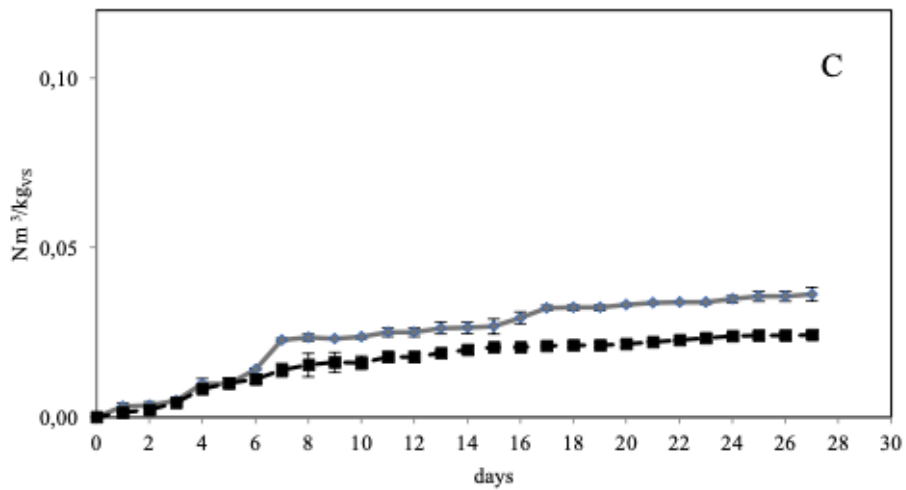
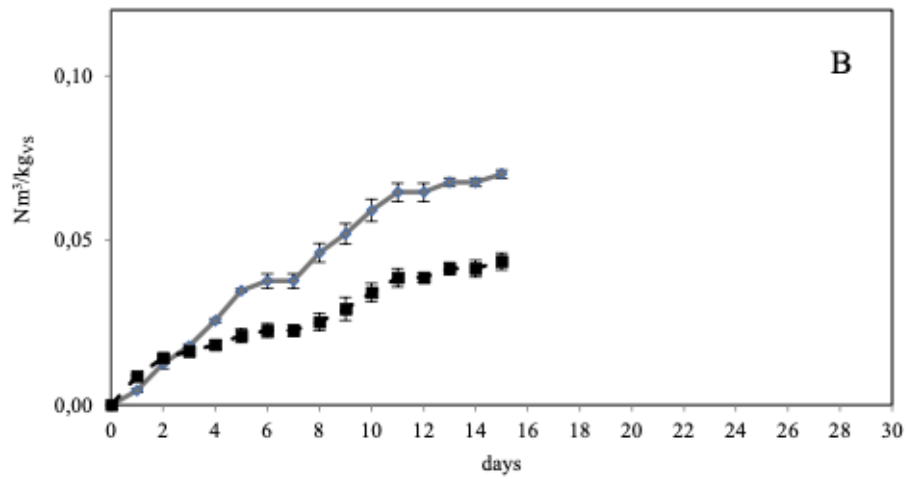
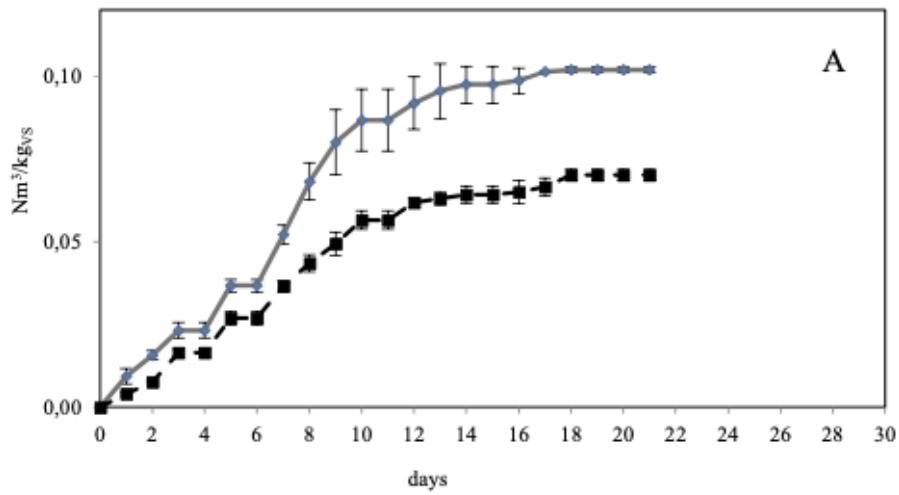
| | sWWTP | RSC | MS | FW | reference |
|---|----------------|-------------|---------------|-------------|--------------------------|
| pH | 6.54±0.49 | 6.30 ±0.17 | 6.69±0.79 | 5.08±0.34 | Demichelis et al., 2018a |
| TS (%) | 2.43±0.74 | 24.98±0.45 | 34.70±2.18 | 25.06 ±3.31 | Demichelis et al., 2018a |
| VS/TS (%) | 86.24±3.2 | 81.65± 1.51 | 75.46±1.28 | 97.17±0.95 | Demichelis et al., 2018a |
| COD _T (mgO ₂ /g vs) | 1174.06 ±94.80 | 1436±23.1 | 1148.52±17.56 | 624.25±48.4 | Demichelis et al., 2018a |
| COD _s /COD _T | 5.56±0.88 | 51.48±1.48 | 6.19±0.26 | 23.24±1.79 | Demichelis et al., 2018a |
| NH ₄ ⁺ [mg/g vs] | 0.29±0.06 | 2.54±0.34 | 0.02±0.01 | 0.02±0.02 | this study |

| | | | | | |
|---------|---|--|---|---|------------|
| C (%) | 44.69±0.57 | 51.60* | 60.50* | 45.65±4.39 | this study |
| H (%) | 7.43±0.16 | 2.05* | 0.30* | 6.76±0.58 | this study |
| N (%) | 4.17±0.44 | 9.40* | 10.30* | 3.24±1.44 | this study |
| S (%) | 1.15±0.24 | 0.90* | 0.11* | 0.05±0.07 | this study |
| formula | C ₁₃ H ₂₆ O ₁₂ N | C ₆ H ₆ O ₆ N | C ₂₄ H ₁₈ O ₁₈ N | C ₂₂ H ₄₀ O ₂₀ N | this study |
| C/N | 11 | 6 | 6 | 14 | this study |

227

228 *3.2. Results of the anaerobic digestion tests performed on the single cosmetic wastes*

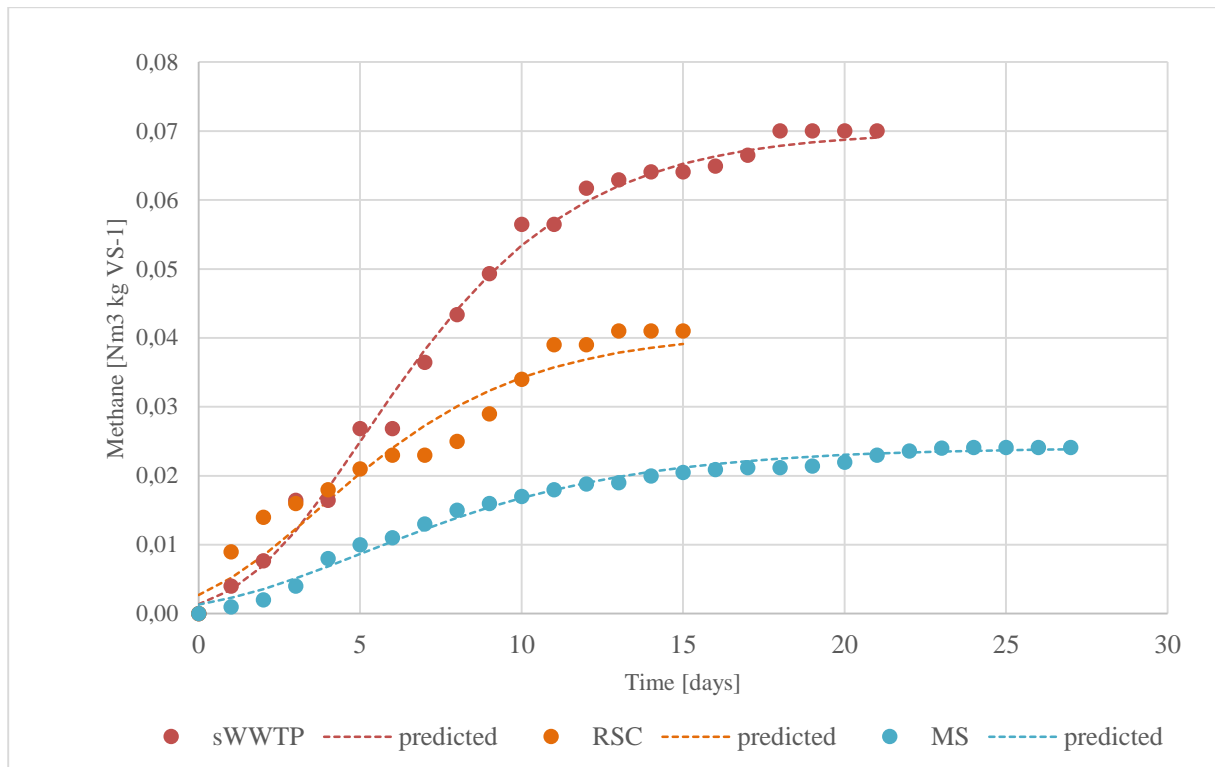
229 The AD tests performed at 35 °C on the single cosmetic wastes (Figure 1) provided some
 230 interesting insights: sWWTP exhibited the highest specific biogas production (0.10 ±0.014
 231 Nm³/kg_{vs}, 70 %-v/v methane), followed by RSC (0.07 ±0.010 Nm³/kg_{vs}, 62 %-v/v methane)
 232 and MS (0.04 ±0.001 Nm³/kg_{vs}, 67 %-v/v methane). These results demonstrated that the
 233 considered cosmetic wastes could undergo AD, even if further actions are needed to improve
 234 their biodegradability and therefore biogas and methane yields. TS removal was 2.1 % for
 235 sWWTP, 24.2 % for RSC and 34.5 % for MS, meaning the following reduction of the amounts
 236 of “raw” (i.e. as they are produced in the plant) wastes: 18.9 %-wt for sWWTP, 3.2 %-wt for
 237 RSC and 1.43 %-wt for MS. As expected from the results of the characterization, sWWTP
 238 showed the highest variability in biogas and methane specific daily productions. It was also
 239 noteworthy the duration of the AD processes: 21 days for sWWTP, 15 days for RSC and 27
 240 days for MS.



241

242 **Figure 1.** Results of AD tests at 35 °C on (A) sWWTP, (B) RSC and (C) MS (specific biogas

243 production in grey; specific methane production in black)



244

245 **Figure 2.** Results of the modeling of experimental methane production from the single cosmetic
 246 wastes at 35 °C through the modified Gompertz model

247

248 **Table 3.** Performance and kinetic parameters of the AD at 35 °C of single cosmetic wastes
 249 obtained through the modified Gompertz model (P: methane potential of the substrate; R_{max} :
 250 maximum methane production rate; λ : lag phase)

| | P (Nm ³ kgVS ⁻¹) | R_{max} (Nm ³ kgVS ⁻¹ d ⁻¹) | λ (days) | R^2 (-) |
|-------|--|--|---------------------|--------------|
| sWWTP | 0.070 | 0.007 | 1.385 | 0.993 |
| RSC | 0.041 | 0.004 | 0.000 | 0.933 |
| MS | 0.024 | 0.002 | 0.302 | 0.986 |

251

252 The performance and kinetic parameters estimated from daily methane production and methane
 253 yield of the single cosmetic wastes at 35 °C through the modified Gompertz model (Table 3,
 254 Figure 2 and Appendix) showed that sWWTP exhibited the highest methane potential
 255 (experimental values) and maximum methane production rate values, compared to RSC and

256 MS, even if with greater lag phase. The fitting of the experimental data through the modified
257 Gompertz model was in all cases satisfactory, achieving coefficient of determination values
258 between 0.93 and 0.99. Comparing the estimated biodegradability of the different waste
259 materials, the highest methane production rate was ascribable, accordingly to the methane
260 potential, in the order sWWTP>RSC>MS; on the contrary, the lag phase length followed the
261 sequence sWWTP>MS>RSC. The highest lag phase estimated for the sWWTP may be due to
262 the already mentioned high concentrations of COD, suspended solids, fats, oils and detergents
263 in the cosmetic industry wastewater (Abidemi et al., 2018) that concentrate in the related waste
264 activated sludge. However, the fact that sWWTP achieved the highest methane potential and
265 production rate values, compared to the other considered samples, demonstrated that this
266 specific waste is anyway biodegradable.

267

268 *3.3. Results of the anaerobic digestion tests performed on mixed cosmetic wastes*

269 The positive effect of the pre-treatments (DR was 64 % \pm 0.01 after TA and 66 % \pm 0.01 after
270 TAS) was appreciated from the results of the AD tests involving the Mix (Table 4 and Figure
271 3). The cumulative biogas/methane production curves showed analogous ultimate yields and
272 different durations of the AD process: 22 days at 35 °C and 17 days at 52 °C, with a faster
273 process in thermophilic conditions, compared to mesophilic, according to literature (among the
274 others, Chi et al., 2010; Ge et al., 2011; Gebreeyessus and Jenicek, 2016).

275

276

277

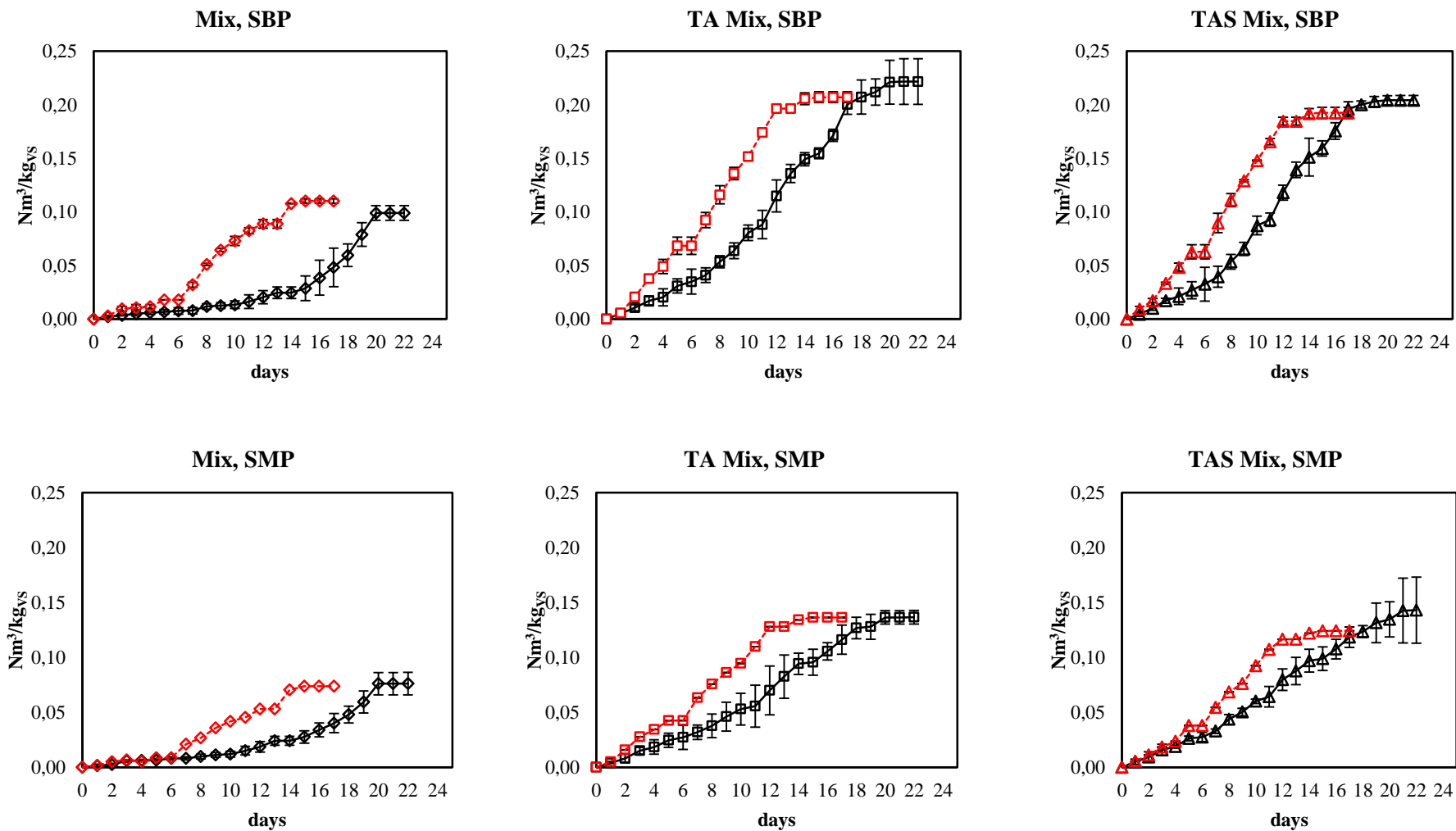
278

279 **Table 4.** Results of the AD tests performed on the Mix at 35 and 52 °C and after TA and TAS
 280 pre-treatments (SBP: specific biogas production; SMP: specific methane production. *only one
 281 replicate was available for SMP at 52 °C)

| | Mix | | TA Mix | | TAS Mix | |
|-----------------------------|-------------|--------------|-------------|--------------|--------------|-------------|
| T (°C) | 35 | 52 | 35 | 52 | 35 | 52 |
| AD duration (d) | 22 | 17 | 22 | 17 | 22 | 17 |
| SBP (Nm ³ /kgvs) | 0.10 ± 0.01 | 0.11 ± 0.002 | 0.22 ± 0.02 | 0.21 ± 0.005 | 0.20 ± 0.004 | 0.19 ± 0.01 |
| SMP (Nm ³ /kgvs) | 0.08 ± 0.01 | 0.07* | 0.14 ± 0.01 | 0.14* | 0.14 ± 0.03 | 0.12* |
| CH ₄ (% v/v) | 77 | 67 | 62 | 66 | 70 | 65 |
| SMP increment (%) | / | / | 75 | 100 | 75 | 71 |

282

283



284 **Figure 3.** Results of AD tests performed at 35 °C (black) and 52 °C (red) on mixed cosmetic wastes (Mix) unaltered and after TA and TAS pre-
 285 treatments (SBP: specific biogas production; SMP: specific methane production). Only one replicate was available for SMP at 52 °C

286 Even if the cosmetic waste materials considered in this work are largely different from any other
 287 substrate already studied, a comparison with state-of-the-art literature related to the application
 288 of pre-treatments to improve AD of sewage sludge (e.g., the most similar substrate in available
 289 literature) could be beneficial anyway. Methane production from sewage sludge resulted
 290 improved after thermal (40 %), alkaline (83 %) and ultrasounds (35-95%) pre-treatments (Zhen
 291 et al., 2017); industrial active sludge exhibited 90 and 104 % methane production increments
 292 after thermal and ultrasound pre-treatments respectively (Carrère et al., 2010).

293

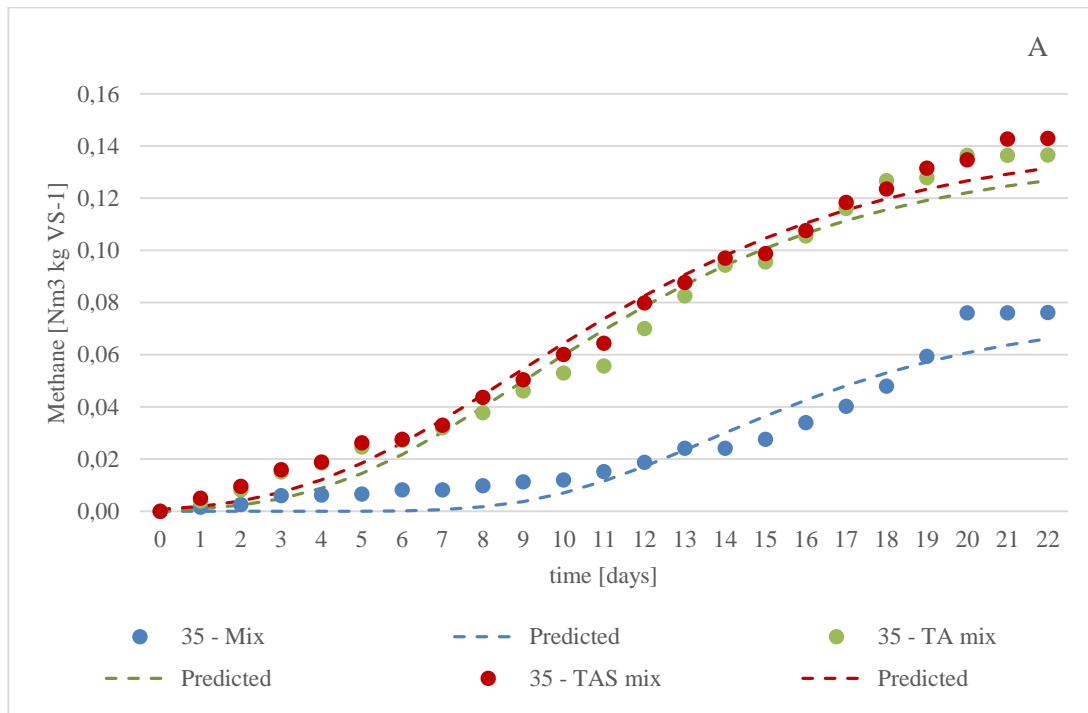
294 **Table 5.** Performance and kinetic parameters of AD at 35 °C and 52 °C of mixed cosmetic
 295 wastes (Mix) before and after TA and TAS pre-treatments obtained through the modified
 296 Gompertz model (P: methane potential of the substrate; R_{max} : maximum methane production
 297 rate; λ : lag phase)

| | P (Nm ³ kgVS ⁻¹) | R_{max} (Nm ³ kgVS ⁻¹ d ⁻¹) | λ (days) | R ² (-) |
|---------|--|--|---------------------|-----------------------|
| 35 °C | | | | |
| Mix | 0.076 | 0.007 | 9.4 | 0.915 |
| TA Mix | 0.137 | 0.010 | 4.0 | 0.972 |
| TAS Mix | 0.143 | 0.010 | 3.5 | 0.983 |
| 52 °C | | | | |
| Mix | 0.074 | 0.009 | 4.8 | 0.975 |
| TA Mix | 0.136 | 0.014 | 2.1 | 0.975 |
| TAS Mix | 0.124 | 0.014 | 2.7 | 0.981 |

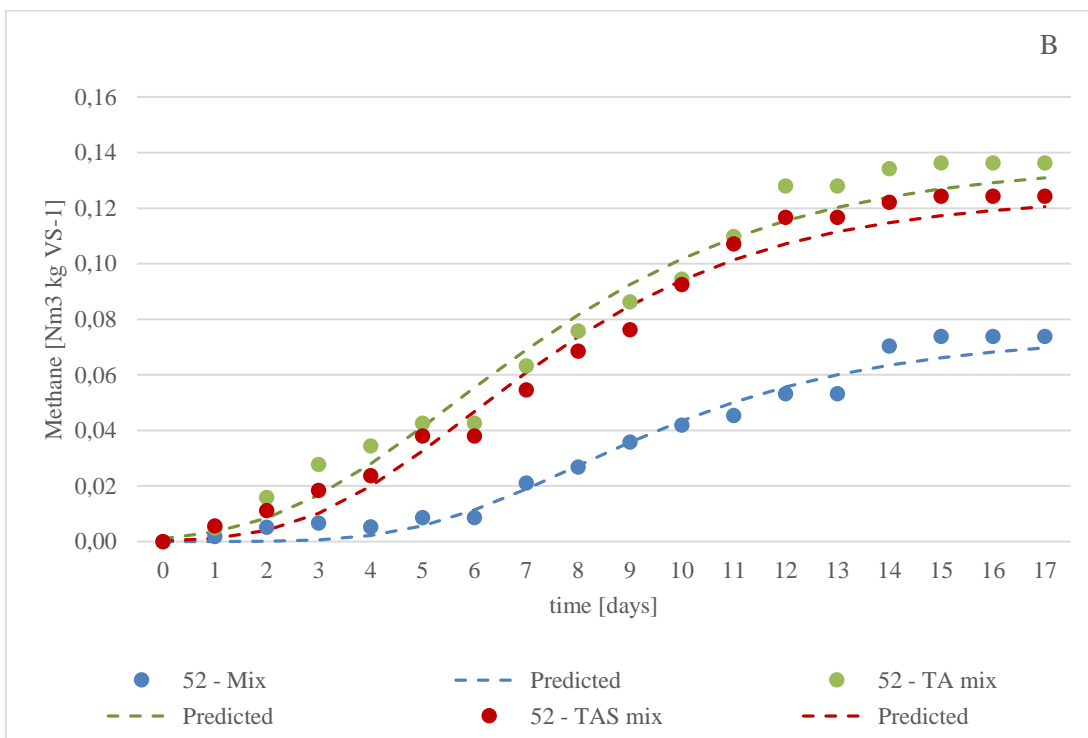
298

299 The performance and kinetic parameters estimated from methane production deriving from the
 300 Mix before and after TA and TAS through the modified Gompertz model (Table 5, Figure 4,
 301 and Appendix) showed that TA and TAS pre-treatments were able to improve the methane
 302 production rate and lag phase. Comparing the pre-treatments, at 35 °C TA Mix exhibited lower
 303 methane potential (experimental values) and equal maximum methane production rate values,

304 compared to TAS Mix, and higher lag phase; at 52°C, TA Mix exhibited higher methane
 305 potential (experimental values) and equal maximum methane production rate values, compared
 306 to TAS Mix, and shorter lag phase. The modified Gompertz model fitting of the experimental
 307 data was in all cases satisfactory, achieving coefficient of determination values between 0.92
 308 and 0.98.



309



310

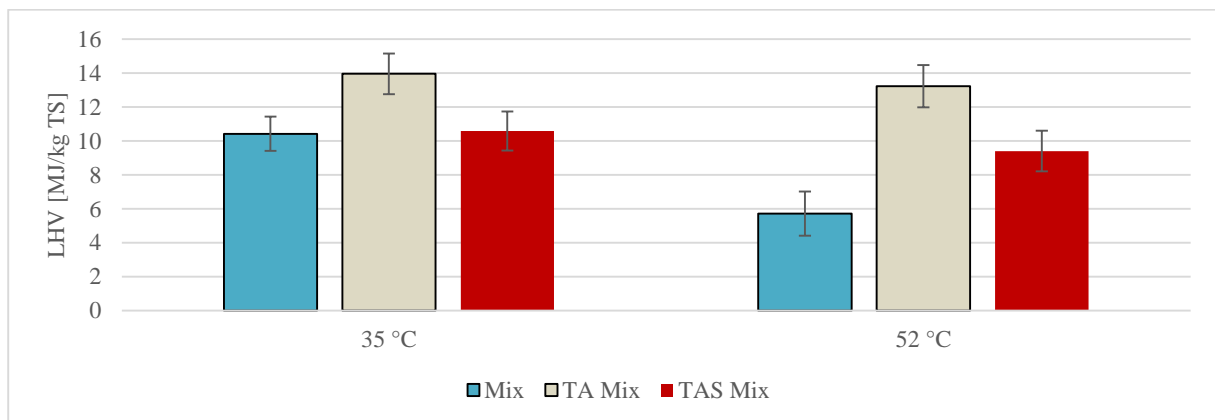
311 **Figure 4.** Results of the modeling of experimental methane production from the mixed cosmetic
312 wastes at 35 (A) and 52 °C (B) through the modified Gompertz model

313 *3.4. Results of digestate characterization*

314 The digestate samples deriving from the AD tests performed on the Mix were characterized
315 with the aim of a preliminary investigation of their thermal valorization. A main critical issue
316 is obviously represented by their high moisture content (95 %-wt), which requires a dehydration
317 treatment before the thermal valorization. The LHV values of the cosmetic waste samples were
318 as follows (values expressed in MJ/kg_{TS}): 20.16±1.01 for sWWTP; 25.97± 1.24 for RSC;
319 25.87±1.30 for MS.

320 Considering the digestates deriving from the Mix, all gave back LHV values similar to wood
321 (12-19 MJ/kg_{TS}) (Green and Perry, 2007). Comparing the LHV values obtained from the Mix
322 after AD at 35 and 52 °C (Figure 5), the thermophilic conditions produced lower LHV values
323 than the mesophilic conditions, in agreement with literature (Gebreyessus and Jenicek, 2016;
324 Khemkhao et al., 2012).

325



326

327 **Figure 5.** LHV values of digestate samples deriving from the AD tests of the Mix at 35 °C and
328 52 °C

329 *3.5. Scale-up and economic assessment*

330 The design parameters (Table 6) adopted in the scale-up analysis (section 2.5) considered AD
 331 in continuous mode, feeding 5 % dry substance, and achieving the performances reported in
 332 section 3.3. The analysis envisioned two scenarios: AD of the Mix “as it was” (S1) and after
 333 TA pre-treatment (S2). Two sub-scenarios, based on the amount of available waste mixture,
 334 were also considered for S2: the amount of waste produced at the moment of the research (a)
 335 and the amount of waste necessary to achieve energy self-sufficiency (b) (section 2.5). The
 336 scale-up assessment based on the amount of waste mixture produced in the plant led for
 337 scenarios S1 and S2.a to a 64 m³ digester, with methane production from the pre-treated Mix in
 338 scenario S2.a (13.7 Nm³/d) almost double than without any pre-treatment in scenario S1 (7.8
 339 Nm³/d). Scenario S2.b led to a 113 m³ digester corresponding to 24.1 Nm³/d methane
 340 production.

341

342 Table 6. Design parameters considered for the scale-up assessment (Mix: waste mixture; TA:
 343 thermo-alkaline pre-treatment with NaOH at 50 °C for 120 min; HRT: hydraulic retention time;
 344 OLR: organic loading rate; Q: flow rate)

| Parameter [measure unit] | S1 (Mix) | S2 (TA Mix) |
|---|----------|-------------|
| TA temperature (T _{TA}) [°C] | - | 50 |
| dose of NaOH [g/g _{TS}] | 0 | 0.08 |
| temperature (T _{AD}) [°C] | 35 | 35 |
| HRT [days] | 22 | 22 |
| Total solids of the mix [kg/m ³] | 140.6 | 140.6 |
| Volatile solids of the mix [kg/m ³] | 117.7 | 117.7 |
| Total solids of the diluted mix [kg/m ³] | 50 | 50 |
| Volatile solids of the diluted mix [kg/m ³] | 41.8 | 41.8 |

| | | | |
|--|-------|-------|------|
| OLR [kgvs/(m ³ day)] | 1.90 | 1.90 | |
| specific methane yield [Nm ³ /kgvs] | 0.080 | 0.140 | |
| Total solids removal [%] | 5.25 | 7.28 | |
| | | a | b |
| Q NaOH [kg/day] | 0 | 9.3 | 16.5 |
| Q Mix [m ³ /d] | 0.8 | 0.8 | 1.5 |
| Q water [m ³ /d] | 1.5 | 1.5 | 2.7 |
| Q diluted Mix [m ³ /d] | 2.3 | 2.3 | 4.1 |
| methane production (Q _{CH₄}) [Nm ³ /day] | 7.8 | 13.7 | 24.1 |
| working volume, 80 % [m ³] | 51 | 51 | 91 |
| volume of the reactor [m ³] | 64 | 64 | 113 |

345

346 The energy analysis (Table 7) showed that in scenarios S1 and S2.a surplus thermal energy was
347 required to heat the digester. In details, considering 303 t/y of waste mixture, TE_{net} was negative,
348 indicating that TE_{CHP} could just partly cover the thermal energy requirements; instead, EE_{net}
349 was positive. The energy surplus needed was larger for the Mix (- 59704 MJ/year) than TA Mix
350 (- 4073 MJ/year). Instead, scenario S2.b was designed to achieve E_{tot} equal to zero, i.e., energy
351 self-sufficiency based on methane production.

352

353 Table 7. Results of energy analysis (Mix: waste mixture; TA: thermo-alkaline pre-treatment
354 with NaOH at 50 °C for 120 min)

| | S1 (Mix) | S2 (TA Mix) | |
|---|----------|-------------|---------|
| | | a | b |
| Net thermal energy (TE _{net}) [MJ/year] | - 92479 | - 67507 | -111786 |
| Heating pre-treatment (TE _{pre}) | 0 | - 5689 | - 10025 |

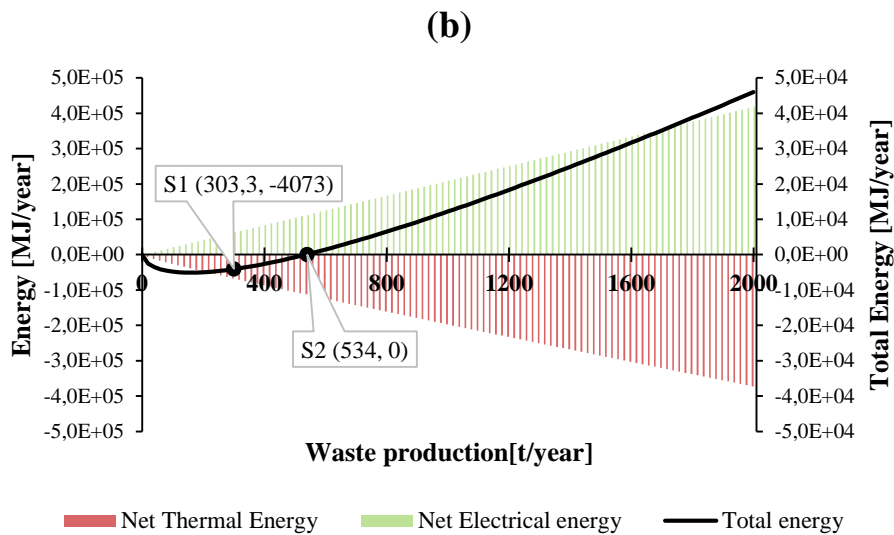
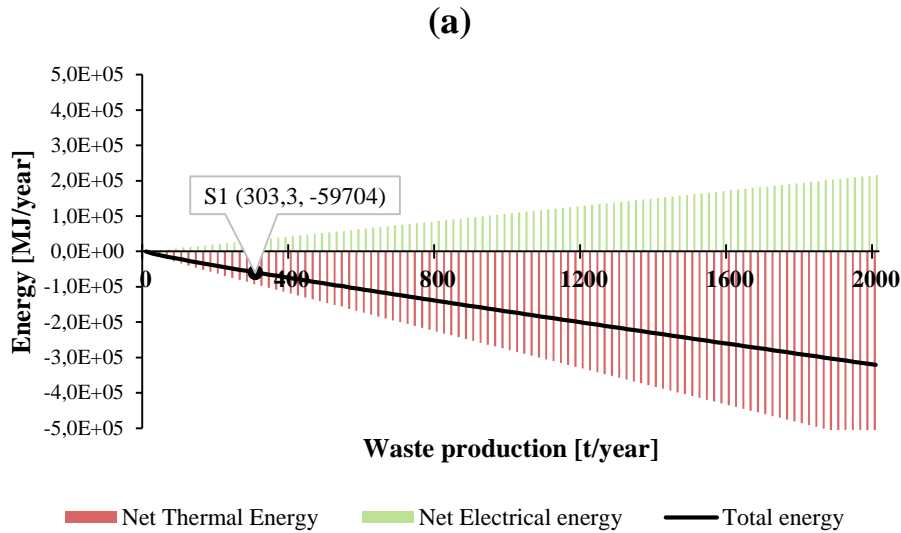
| | | | |
|--|----------|----------|----------|
| Heating feeding AD (TE_{AD}) | - 109693 | - 109693 | - 193305 |
| Thermal loss AD (TE_{loss}) | - 23665 | - 23665 | - 34527 |
| Thermal energy from CHP (TE_{CHP}) | + 40880 | + 71540 | + 126071 |
| <hr/> | | | |
| Net electrical energy (EE_{net}) [MJ/year] | + 32774 | + 63434 | + 111786 |
| <hr/> | | | |
| Electricity mixing AD (EE_{mix}) | - 8106 | - 8106 | - 14284 |
| Electricity from CHP (EE_{CHP}) | + 40880 | + 71540 | + 126071 |
| <hr/> | | | |
| Total energy (E_{tot}) [MJ/year] | - 59704 | - 4073 | 0 |
| <hr/> | | | |

355

356 Despite the waste production at the moment of the research was not sufficient to reach the
357 energetic self-sustainability of AD at 35 °C, TA pre-treatment could be the key towards the
358 process energetic sustainability by significantly enhancing the specific methane yield (from
359 0.080 Nm³/kg_{vs} to 0.140 Nm³/kg_{vs}). In fact, investigating the energy demand as a function of
360 the amount of waste mixture undergoing AD at 35 °C (Figure 6), E_{tot} was negative for any
361 values of waste production (Figure 6a), and E_{tot} even decreased by enhancing the waste
362 production. Therefore, the energetic self-sustainability couldn't be reached for without TA pre-
363 treatment. Instead, in case of the AD of the TA mix at 35 °C (Figure 6b), E_{net} was positive for
364 waste production larger than 534 t/year.

365

366 Figure 6. Total energy, net thermal energy, and net electrical energy as a function of the waste
367 production: (a) AD at 35 °C of the Mix, (b) AD at 35 °C of the TA pre-treated Mix.



368
369

370 The results and details of the economic analysis (Table 8) showed that both the Mix and TA
 371 Mix options could be profitable with the current waste production (scenarios S1 and S2.a), with
 372 promising NPV and ROI of 108,694 € and 13.1 % for the Mix, and 112,936 € and 13.3 % for
 373 TA Mix. In both cases the payback time of 10 years was reasonable. Despite both scenarios
 374 didn't reach the energetic sustainability (natural gas was hypothesized as thermal energy
 375 source), they showed promising economic performances. This is not surprising, since the major
 376 contribution to the total revenues (Table 8) didn't come from the sale of electricity (representing
 377 4.5 % of total revenues for the Mix and 7.6 % for TA Mix), but from the savings related to the

378 disposal of the digestate compared to the current waste management (95.5 % of total revenues
 379 for the Mix and 92.4 % for TA Mix). Considering S2.b, the profitability was larger than in other
 380 scenarios, with NPV of 270,294 €, ROI of 15.2 % and payback time of 9 years.

381

382 Table 8. Results of the economic analysis: operational costs, revenues, total investment costs,
 383 return of investment (ROI), net present value (NPV), payback time (PBT).

| | S1(Mix) | S2 (TA mi) | | Notes | References |
|--|---------|------------|-------|---|--------------------------------|
| | | a | b | | |
| Total | 9182 | 9910 | 11744 | | |
| operational costs [€/year] | | | | | |
| NaOH [€/year] | 0 | 921 | 1623 | NaOH: cost 0.27 €/kg; dose 0.08 g/gTS | Demichelis et al., 2018a |
| water [€/year] | 753 | 753 | 1327 | water cost: 1.37 €/m ³ | Demichelis et al., 2018a |
| Net thermal energy [€/year] | 717 | 523 | 866 | non-household consumers - natural gas cost average EU-27 (2 nd half 2020): 0.0279 €/kWh | Eurostat, 2021 |
| Electricity [€/year] | 282 | 282 | 498 | non-household consumers - electricity cost average EU-27 (2 nd half 2020): 0.1254 €/kWh | Eurostat, 2021 |
| Labour [€/year] | 7430 | 7430 | 7430 | labour cost average EU-27: 28.5 €/hour; 1 worker; 1 hours/day; 5 days/week | Eurostat, 2021 |
| Total revenues [€/year] | 31633 | 32701 | 57626 | electricity cost average EU-27 (2 nd half 2020): 0.1254 €/kWh | Eurostat, 2021 |
| Save costs for waste disposal [€/year] | 30209 | 30209 | 53235 | waste disposal: sWWTP: 0.15 €/kg; RSC and MS: 0.31 €/kg; FW: 0.110 €/kg; digestate: 0.120 €/kg | Demichelis et al., 2018a |

| | | | | | |
|------------------------------|--------|--------|--------|---|---------------------------|
| Electricity [€/year] | 1424 | 2492 | 4391 | electricity cost average EU-27 (2 nd half 2020): 0.1254 €/kWh | Eurostat, 2021 |
| Total investment cost [€] | 171087 | 171087 | 301495 | FCC: Cost digester construction: 2500 €/m ³ WCC: 6.5 % of FCC | Chiappero et al., 2019 |
| ROI [%] | 13.1 | 13.3 | 15.2 | | Chiappero et al., 2019 |
| NPV [€] | 108694 | 112936 | 270294 | lifetime (n): 20 years; discount rate (i): 5 % | Chiappero et al., 2019 |
| PBT [years] | 10 | 10 | 9 | | Chiappero et al., 2019 |

384

385 4. Conclusions

386 The management of the industrial wastes deriving from cosmetic products manufacturing is
387 challenging from the technical and economic viewpoints: wastewater, rich of scarcely
388 biodegradable organic compounds, is typically treated in onsite treatment plants that are highly
389 efficient in COD reduction; other wastes, including the wastewater sludge, are usually landfilled
390 or incinerated. While the available literature on cosmetic waste management only offers few
391 data on the AD of wastewater, this work was aimed at investigating the technical feasibility of
392 the AD of different cosmetic wastes deriving from one of the largest plants of L'Oréal Group
393 in Europe.

394 A first set of AD tests at 35 °C concerning the single waste materials provided encouraging
395 results (up to 0.10 Nm³/kg_{vs} of biogas, 70 %-v/v methane). Further AD tests involved a mixture
396 (according to the amounts produced in the plant) of the wastes at 35 and 52 °C. Considering the
397 low ratio between soluble and total COD, the wastes underwent physico-chemical pre-
398 treatments (thermo-alkaline and thermo-alkaline-sonication). The main findings of this research
399 were as follows: pre-treatments considerably increased (up to 100 %) methane yields compared

400 to untreated mixed wastes, with better performances for the thermo-alkaline pre-treatment; no
401 significant differences were observed comparing the performances of the AD of pre-treated
402 mixed wastes in mesophilic and thermophilic conditions, achieving in all cases biogas yields
403 around 0.2 Nm³/kg_{VS} and 62-70 % methane; after AD, 5-8 %-wt total solids reductions were
404 observed; even if a dewatering process will be necessary, the thermal valorization of the
405 digestate could be investigated in the future.

406 On the grounds of the technical and economic assessment of the feasibility of the scale-up of
407 the overall process, TA pre-treatment exhibited a key role in achieving the process energetic
408 sustainability. The economic analysis showed that with the current waste production AD of the
409 considered cosmetic waste could be sustainable. In fact, even if the energetic sustainability
410 wasn't achieved (natural gas was hypothesized as thermal energy source), the major
411 contribution to the total revenues derived from the savings related to the disposal of the digestate
412 compared to the current waste management. In conclusion, considering the lack of literature on
413 the anaerobic digestion of cosmetic wastes, this study would like to provide an insight on the
414 topic and stimulate further exploration of alternative management options for cosmetic wastes.

415

416 **Acknowledgements**

417 The authors declare no conflict of interest. The Authors gratefully acknowledge L'Oréal Saipo
418 Industriale SpA for the financial support to the research in 2016 and for approving the
419 submission of this manuscript. The Authors acknowledge SMAT and ACEA Pinerolese for
420 supplying the digestate samples employed as inoculum in the AD tests. Authors' contributions
421 are detailed in the following. S. Fiore: conceptualization, methodology, supervision, data
422 curation, writing-original draft writing and manuscript review, funding acquisition; F.
423 Demichelis: experimental activity, support to original draft writing; M. Chiappero: data curation

424 and writing-original draft writing and manuscript review; M. Onofrio: conceptualization,
425 methodology and writing-manuscript review.

426

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