

# Techno-Economic and Environmental Analysis of a Sewage Sludge Alternative Treatment Combining Chemical Looping Combustion and a Power-to-Methane System

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## Supplementary Material

## **Nomenclature**

$a$	char surface-to-volume ratio, $\text{m}^2/\text{m}^3$
$c_{p,i}$	specific heat capacity, $\text{J}/(\text{mol K})$
$d$	Sauter mean particle diameter, mm
$E_C$	amount of carbon fines elutriated from the fuel reactor, kg/s
$[i]$	gas phase concentration, $\text{mol}/\text{m}^3$
$\dot{m}_{char}^{in CS}$	inlet char flow rate from the fuel reactor to the carbon stripper, kg/s
$\dot{n}_i$	molar flow rate, mol/s
$P_i$	partial pressure, Pa
$\dot{Q}$	thermal power, kW
$R$	gas constant, $\text{J}/(\text{K mol})$
$r_j$	reaction rate, mol/s
$S_{FB}$	cross-section of the fuel reactor, $\text{m}^2$
$T$	absolute temperature, K
$u$	gas superficial velocity, m/s
$W_i$	inventory in the fuel reactor, kg
$X_{CH_4}$	methane conversion degree, -
$X_{char}$	char conversion degree, -
$y_i$	gaseous molar fraction, -

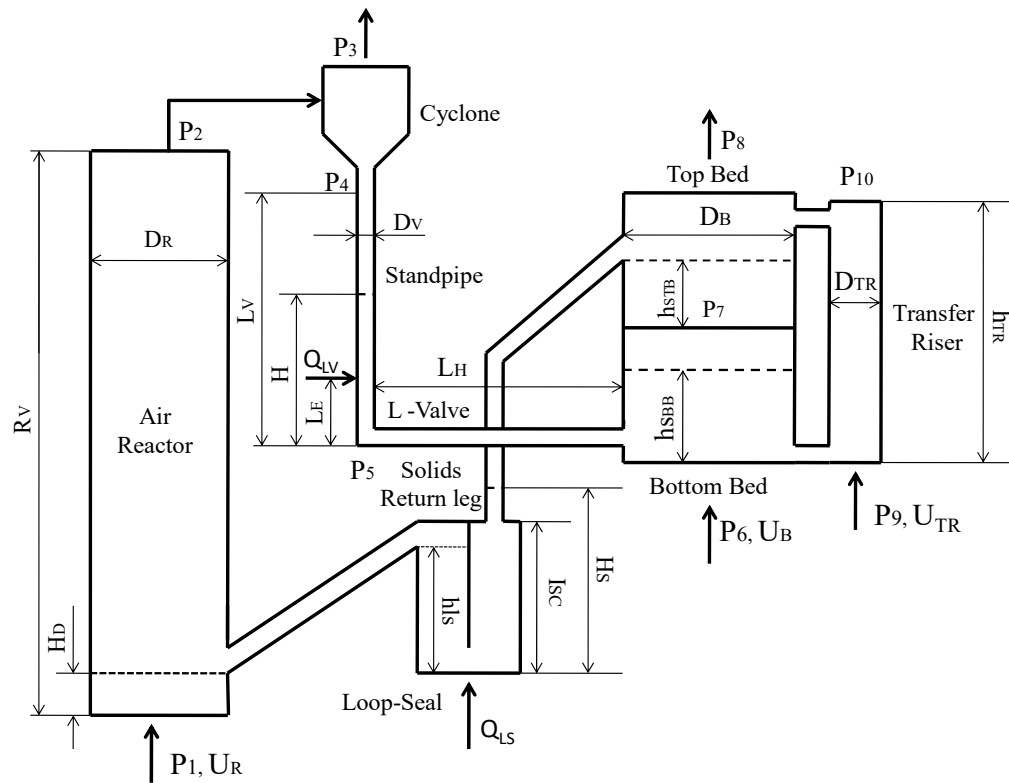
## *Greek letters*

$\alpha$	stoichiometric coefficient, -
$\Delta H_{r_j}^0$	standard enthalpy of reaction, J/mol
$\eta_{CC}$	CO <sub>2</sub> capture efficiency, -
$\eta_{CO_2}$	carbon-to-CO <sub>2</sub> efficiency, -
$\eta_{CS}$	efficiency of the carbon stripper, -
$\eta_{CS}^{max}$	maximum efficiency of the carbon stripper, -
$\eta_{FR}$	combustion efficiency, -
$\rho_C$	char density, $\text{kg}/\text{m}^3$
$\Phi$	oxygen-to-fuel ratio, -
$\psi$	Hurt's kinetic parameter, -

## *Subscripts or superscripts*

$0$	standard state
$AR$	air reactor
$D$	dense zone in the fuel reactor
$El$	elutriated material
$FB$	freeboard
$FR$	fuel reactor
$S$	oxygen carrier support
$in$	incoming
$in - V$	incoming volatile matter
$mf$	incipient fluidization
$out$	outcoming

Fig. 1 SM reports the multiple interconnected fluidized (MFB) bed system modelled. The MIF reactor is equipped with a two-stage fuel reactor, a riser used as Air Reactor, a cyclone, a L-valve return leg, and a loop-seal.



**Figure S1.** Schematic representation of the MFB system along with the indication of main geometric and operating variables.

## S1 Hydrodynamic model.

### Riser

Two different operating regimes are considered: i) dilute regime, in which pneumatic transport of solids along the riser is established; ii) dense regime, that is characterized by the presence of a dilute phase above a dense bed.

In the dense flow regime, the correlation proposed by Wirth has been used for the solid mass flux  $G_{WIRTH}$  at saturation:

$$G_{WIRTH} = \beta \rho_p (1 - \varepsilon_{mf}) U_R \quad (SI.1)$$

$$\beta = \left[ \left( 1 - \frac{U_t}{U_R} \right)^2 0.00533 \frac{Fr^2}{4(1 - \varepsilon_{mf})} \right] \eta^* (U_R - U_t) \quad (SI.2)$$

While, in dilute regime, the solid max flux,  $G_{diluted}$ , is:

$$G_{diluted} = \frac{1}{A_R} \cdot \left( \frac{U_s}{h_R} \right) \cdot m_R \quad (SI.3)$$

The solid mass flux,  $G_s$ , at the outlet of riser is hence defined as:

$$G_s = \begin{cases} G_{diluted} & \rightarrow G_s < G_{WIRTH} \\ G_{WIRTH} & \rightarrow G_s \geq G_{WIRTH} \end{cases} \quad (SI.3)$$

The gas pressure drop along the riser is assumed to be due only to the contribution of gravitational force:

$$\Delta P_R = \rho_p (1 - \varepsilon_D) g h_D + \frac{g W_R}{A_R U_S} (h_R - h_D) \quad (SI.4)$$

### *Cyclone*

For the cyclone, the collection efficiency was assumed to be 1. The gas pressure drop is evaluated according to:

$$\Delta P_{cyc} = \rho_f K_c U_c \quad (SI.5)$$

### *Downcomer/L-Valve*

The pressure drop in the downcomer is evaluated according to:

$$\frac{\Delta P_{DOW}}{H - L_E} = K_V (u_{fy} - u_{sy}) \quad (\text{SI.6})$$

$$u_{fy} = \frac{4Q_V}{\pi D_{LV}^2 \varepsilon_V} \quad (\text{SI.7})$$

$$u_{sy} = \frac{-4W_S}{\pi D_{LV}^2 (1 - \varepsilon_V) \rho_P} \quad (\text{SI.8})$$

The same approach is used to evaluated the pressure drop in the L-Valve, considering that in the horizontal passage of L-valve, the pressure drop is due to viscous friction between gaseous and solid phase, as expressed by the Lewis equation:

$$\frac{\Delta P_{LV}}{L_H} = K_H (u_{fx} - u_{sx}) \quad (\text{SI.9})$$

$$u_{fx} = \frac{4Q_H}{\pi D_{LV}^2 \varepsilon_H} \quad (\text{SI.10})$$

$$u_{sx} = \frac{-4W_S}{\pi D_{LV}^2 (1 - \varepsilon_H) \rho_P} \quad (\text{SI.11})$$

At the same time, it can be described according to:

$$\Delta P_{LV} = \frac{0.0649 \rho_P^{0.996} L_H}{D_{LV}^{0.574} d_P^{0.237}} \left( \frac{W_S}{A_R} \right)^{0.178} \quad (\text{SI.12})$$

Moreover, it was taken into account that the aeration gas flow rate in the L-valve is divided into a vertical gas flow rate along the downcomer and the horizontal gas flow rate along the L-valve:

$$Q_{LV} = Q_H + Q_V \quad (\text{SI.13})$$

### *Loop-Seal*

For the sake of simplicity, the loop-seal is modelled as a zero-order node: it supplied to the riser the same mass flow rate discharged into the BFB by the L-valve. To avoid the

formation of potential explosive O<sub>2</sub>-H<sub>2</sub> mixture, the “leakage” of gas from the supply chamber to the recycle chamber is prevented taking into account the relationship:

$$U_{LS} \geq U_{mf} - \frac{W_R \varepsilon_{mf}}{A_{SC} \rho_p (1 - \varepsilon_{mf})} \quad (\text{SI.14})$$

Moreover, aeration gas flow rate in the loop-seal was assumed to be equally divided into supply and recycle chamber.

*Global balance and dynamic model*

$$m_R = \frac{A_R}{g} \cdot \Delta P_R \quad (\text{SI.15})$$

$$m_{D/LV} = A_H \cdot \rho_p \cdot (1 - \varepsilon_H) \cdot L_H + \rho_p \cdot (1 - \varepsilon_V) \cdot A_D \cdot [H \cdot \eta (L_V - H) + L_V \cdot \eta^* (H - L_V)] \quad (\text{SI.16})$$

$$m_{BFB} = \frac{A_B}{g} \cdot \Delta P_{BFB} \quad (\text{SI.17})$$

$$m_{LS} = (1 - \varepsilon_m) \cdot \rho_p \cdot A_{SP} \cdot (L_S - I_{SC}) + (1 - \varepsilon_m) \cdot \rho_p \cdot A_{SC} \cdot I_{SC} + (1 - \varepsilon_R) \cdot \rho_p \cdot A_{RC} \cdot h_{RC} \quad (\text{SI.18})$$

where  $\eta$  and  $\eta^*$  are:

$$\begin{aligned} \eta(x) &= \begin{cases} 0 \rightarrow x < 0 \\ 1 \rightarrow x \geq 0 \end{cases} \\ \eta^*(x) &= \begin{cases} 0 \rightarrow x \leq 0 \\ 1 \rightarrow x > 0 \end{cases} \end{aligned} \quad (\text{SI.19})$$

As regards the bed voidage in the loop seal,  $\varepsilon_R$  is assumed equal to 0.5 while  $\varepsilon_m$  was evaluated as:

$$\varepsilon_m = 1 - \frac{1 - \varepsilon_0}{R_l} \quad (\text{SI.20})$$

$$R_1 = \begin{cases} 3.1 \cdot (d_p \cdot \rho_p)^{0.5} & \rightarrow \frac{u_0}{U_t} \leq 0.0168 \cdot \left( \frac{u_0}{U_t} \right)^{-0.6} \\ 23.94 \cdot (d_p \cdot \rho_p)^{0.3} \cdot \frac{u_0}{U_t} & \rightarrow \frac{u_0}{U_t} > 0.0168 \cdot \left( \frac{u_0}{U_t} \right)^{-0.6} \end{cases} \quad (\text{SI.21})$$

$$u_0 = \frac{4 \cdot Q_{sc}}{\pi D_{sp}^2} \quad (\text{SI.22})$$

## S2 Kinetic model

The model is based on the following hypothesis:

- 1) steady-state;
- 2) the dense zone of fuel reactor is well stirred with respect to both gas and solid species;
- 3) the freeboard of fuel reactor conforms to plug flow;
- 4) if air reactor works in diluted regime, it is well stirred respect to both gas and solid species; instead, if air reactor works in dense regime, it conforms to plug flow;
- 5) conversion is uniform throughout the solid particles;
- 6) steam and dry-reforming are negligible in the freeboard, in which only water gas shift reaction is take into account;
- 7) fuel reactor is isothermal;
- 8) air reactor is adiabatic.

Model equations consist of mass and energy balances on reagents and products written for both air and fuel reactors.

The mass balance equations, specialized to model the dense phase for both the bottom and the top bed, were referred or to single elements (i.e. C, H, O, Cu) or to specific compounds (i.e. CuO, char, O<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>O) assuming the following general form:

$$\sum \dot{n}_i^{in} - \sum \dot{n}_i^{out} + \sum (\alpha_{i,j} \cdot r_j) = 0 \quad (\text{SI.23})$$

The complete set of equations is reported in Table S1

Table S1. Mass balance equations for the dense zone (bottom and top bed) in the fuel reactor

Element or compound	Equation
Atomic carbon	$\dot{n}_C^{in} + \dot{n}_{CO_2}^{in} + \dot{n}_{CO}^{in} + \dot{n}_{CH_4}^{in} - \dot{n}_C^{out} - \dot{n}_{gas}^{out} \cdot y_{CO_2}^{out} - \dot{n}_{gas}^{out} \cdot y_{CO}^{out} - \dot{n}_{gas}^{out} \cdot y_{CH_4}^{out} = 0$
Atomic copper	$\dot{n}_{CuO}^{in} + 2\dot{n}_{Cu_2O}^{in} + \dot{n}_{Cu}^{in} - \dot{n}_{CuO}^{out} - 2\dot{n}_{Cu_2O}^{out} - \dot{n}_{Cu}^{out} = 0$
Atomic hydrogen	$\dot{n}_{H_2O}^{in} + \dot{n}_{H_2}^{in} + 2\dot{n}_{CH_4}^{in} - \dot{n}_{gas}^{out} \cdot y_{H_2O}^{out} - \dot{n}_{gas}^{out} \cdot y_{H_2}^{out} - 2\dot{n}_{gas}^{out} \cdot y_{CH_4}^{out} = 0$
Atomic oxygen	$\dot{n}_{CuO}^{in} + \dot{n}_{Cu_2O}^{in} + 2\dot{n}_{CO_2}^{in} + \dot{n}_{CO}^{in} + 2\dot{n}_{O_2}^{in} + \dot{n}_{H_2O}^{in} - \dot{n}_{CuO}^{out} - \dot{n}_{Cu_2O}^{out} - 2\dot{n}_{gas}^{out} \cdot y_{O_2}^{out} - 2\dot{n}_{gas}^{out} \cdot y_{CO_2}^{out} - \dot{n}_{gas}^{out} \cdot y_{H_2O}^{out} - \dot{n}_{gas}^{out} \cdot y_{CO}^{out} = 0$
copper oxide (II)	$\dot{n}_{CuO}^{in} - \dot{n}_{CuO}^{out} - W_{CuO} \cdot (r_2 + r_4 + r_9 + r_{11}) = 0$
copper	$\dot{n}_{Cu}^{in} - \dot{n}_{Cu}^{out} + 2W_{Cu_2O} \cdot (r_3 + r_{10}) + W_{CuO} \cdot r_{11} = 0$
carbon (char)	$\dot{n}_C^{in} - \dot{n}_C^{out} + W_C \cdot (-r_1 - r_6 - r_7) = 0$
Molecular oxygen	$\dot{n}_{O_2}^{in} - \dot{n}_{gas}^{out} \cdot y_{O_2}^{out} + \frac{1}{4}W_{CuO} \cdot r_4 - W_C \cdot r_6 - \frac{1}{2}V^D \cdot (r_5 + r_8) - 2V^D \cdot r_{12} = 0$
water	$\dot{n}_{H_2O}^{in} - \dot{n}_{gas}^{out} \cdot y_{H_2O}^{out} - W_C \cdot r_7 + V^D \cdot r_8 + \frac{1}{2}W_{CuO} \cdot (r_9 + r_{11}) + W_{Cu_2O} \cdot r_{10} + 2V^D \cdot r_{12} = 0$
methane	$\dot{n}_{CH_4}^{in} - \dot{n}_{gas}^{out} \cdot y_{CH_4}^{out} - \frac{1}{4}W_{CuO} \cdot r_{11} - V^D \cdot r_{12} = 0$
Gaseous species	$1 - y_{CO_2}^{out} - y_{CO}^{out} - y_{O_2}^{out} - y_{H_2O}^{out} - y_{H_2}^{out} - y_{CH_4}^{out} = 0$

In the freeboard of the bottom and the top bed, where only homogeneous reactions of the gaseous compounds were taken into account and the complete set of equations is reported in

Table S2

Table S2. Differential equations for mass balance in the freeboard of the fuel reactor

Compound	Equation
Carbon dioxide	$\frac{1}{S_{FB}} \frac{d\dot{n}_{CO_2}^{FB}}{dh} = r_5 + r_{12}$
Carbon monoxide	$\frac{1}{S_{FB}} \frac{d\dot{n}_{CO}^{FB}}{dh} = -r_5$
Oxygen	$\frac{1}{S_{FB}} \frac{d\dot{n}_{O_2}^{FB}}{dh} = -\frac{1}{2}r_5 - \frac{1}{2}r_8 - 2r_{12}$



Water	$\frac{1}{S_{FB}} \frac{d\dot{n}_{H_2O}^{FB}}{dh} = r_8 + 2r_{12}$
Hydrogen	$\frac{1}{S_{FB}} \frac{d\dot{n}_{H_2}^{FB}}{dh} = -r_8$
Methane	$\frac{1}{S_{FB}} \frac{d\dot{n}_{CH_4}^{FB}}{dh} = -r_{12}$
Total gas moles	$\frac{1}{S_{FB}} \frac{d\dot{n}_{gas}^{FB}}{dh} = -\frac{1}{2}r_5 - \frac{1}{2}r_8$

Energy balance equations, used to calculate the thermal power needed to ensure isothermal reactor operation at the pre-set temperature and the detailed equations are reported in Table S3.

Table S3. Energy balance equations for the fuel and air reactor.

Description	Equation
Fuel Reactor energy balance	$ \begin{aligned} & (\dot{n}_S^{in} c_{p,S} + \dot{n}_{CuO}^{in} c_{p,CuO}) \cdot (T^0 - T^{AR}) + \dot{n}_{gas}^{in} \\ & \cdot (y_{CO_2}^{in} c_{p,CO_2} + y_{H_2O}^{in} c_{p,H_2O}) \cdot (T^0 - T_{gas}^{in}) \\ & + (\dot{n}_C^{in} c_{p,C} + \dot{n}_{V,H_2O}^{in} c_{p,H_2O(l)} + \dot{n}_{V,CO}^{in} c_{p,CO} + \dot{n}_{V,H_2}^{in} c_{p,H_2} \\ & + \dot{n}_{V,CH_4}^{in} c_{p,CH_4} + \dot{n}_{Ash}^{in} c_{p,Ash}) \cdot (T^0 - T_{Fuel}^{in}) + \dot{n}_{V,H_2O}^{in} \\ & \cdot (-\lambda_{vap,H_2O}) \\ & + [\dot{n}_S^{out} c_{p,S} + \dot{n}_{CuO}^{out} c_{p,CuO} + \dot{n}_{Cu_2O}^{out} c_{p,Cu_2O} + \dot{n}_{Cu}^{out} c_{p,Cu} \\ & + \dot{n}_C^{out} c_{p,C} + \dot{n}_{Ash}^{out} c_{p,Ash} + \dot{n}_{gas}^{out} \\ & \cdot (y_{CO_2}^{out} c_{CO_2} + y_{CO}^{out} c_{CO} + y_{O_2}^{out} c_{O_2} + y_{H_2O}^{out} c_{H_2O} + y_{H_2}^{out} c_{H_2} \\ & + y_{CH_4}^{out} c_{CH_4})] \cdot (T^{FR} - T^0) + W_C \cdot (-\Delta H_{r1}^0) \cdot r_1 + W_{CuO} \\ & \cdot (-\Delta H_{r2}^0) \cdot r_2 + W_{Cu_2O} \cdot (-\Delta H_{r3}^0) \cdot r_3 + W_{CuO} \\ & \cdot (-\Delta H_{r4}^0) \cdot r_4 + V^{FR} \cdot (-\Delta H_{r5}^0) \cdot r_5 + W_C \cdot (-\Delta H_{r6}^0) \cdot r_6 \\ & + W_C \cdot (-\Delta H_{r7}^0) \cdot r_7 + V^{FR} \cdot (-\Delta H_{r8}^0) \cdot r_8 + W_{CuO} \\ & \cdot (-\Delta H_{r9}^0) \cdot r_9 + W_{Cu_2O} \cdot (-\Delta H_{r10}^0) \cdot r_{10} + W_{CuO} \\ & \cdot (-\Delta H_{r11}^0) \cdot r_{11} + V^{FR} \cdot (-\Delta H_{r12}^0) \cdot r_{12} + \dot{Q}^{FR} = 0 \end{aligned} $
Air Reactor energy balance	$ \begin{aligned} & (\dot{n}_S^{in} c_{p,S} + \dot{n}_{CuO}^{in} c_{p,CuO} + \dot{n}_{Cu_2O}^{in} c_{p,Cu_2O} + \dot{n}_{Cu}^{in} c_{p,Cu} + \dot{n}_C^{in} c_{p,C} \\ & + \dot{n}_{Ash}^{in} c_{p,Ash}) \cdot (T^0 - T^{FR}) + \dot{n}_{air}^{in} \cdot [y_{O_2}^{out} c_{O_2} + y_{N_2}^{out} c_{N_2}] \\ & \cdot (T^0 - T^{air}) \\ & + [\dot{n}_S^{out} c_{p,S} + \dot{n}_{CuO}^{out} c_{p,CuO} + \dot{n}_{Ash}^{out} c_{p,Ash} + \dot{n}_{gas}^{out} \\ & \cdot (y_{CO_2}^{out} c_{CO_2} + y_{O_2}^{out} c_{O_2} + y_{N_2}^{out} c_{N_2})] \cdot (T^{AR} - T^0) + \dot{n}_C^{in} \\ & \cdot (-\Delta H_{r6}^0) + \dot{n}_{Cu}^{in} \cdot (-\Delta H_{r13}^0) + \dot{n}_{Cu_2O}^{in} \cdot (-\Delta H_{r14}^0) + \dot{Q}^{AR} \\ & = 0 \end{aligned} $

The kinetic expressions adopted in the model are reported in Table S4. For the reactions R13 and R14 no kinetic expression were needed as they were considered complete in the air reactor.

Table S4. Kinetic reaction rates

kinetic expression	unit
$r_1 = 1.09 * 10^9 * (P_{CO_2})^{0.54} * e^{\left[ \frac{283 \frac{kJ}{mol}}{RT} \right]} * (1 - X_{char}) * [1 - \psi * \ln(1 - X_{char})]^{0.5}$	$\frac{kgC}{kgC \cdot s}$
$r_2 = 2.83 * 10^7 * e^{\left[ \frac{50 \frac{kJ}{mol}}{RT} \right]} * [CO]$	$\frac{mol CO}{m^3 \cdot s}$
$r_3 = 1.49 * 10^7 * e^{\left[ \frac{45 \frac{kJ}{mol}}{RT} \right]} * [CO]$	$\frac{mol CO}{m^3 \cdot s}$
$r_4 = 49.4 * e^{-\frac{110 \frac{kJ}{mol}}{RT}}$	$\frac{gO_2}{gCu \cdot s}$
$r_5 = 1.3 * 10^8 * e^{\left[ \frac{-15106 K}{T} \right]} * [CO] * [O_2]^{0.5} * [H_2O]^{0.5}$	$\frac{mol CO}{m^3 \cdot s}$
$r_6 = 2.902 * 10^3 * \frac{a}{\rho_C} * e^{\left[ \frac{85639 \frac{kJ}{mol}}{RT} \right]} * P_{O_2}$	$\frac{mol C}{mol C \cdot s}$
$r_7 = 2.47 * 10^7 * (P_{H_2O})^{0.86} * e^{\left[ \frac{214 \frac{kJ}{mol}}{RT} \right]} * (1 - X_{char}) * [1 - \psi * \ln(1 - X_{char})]^{0.5}$	$\frac{kgC}{kgC \cdot s}$
$r_8 = \frac{3.26 * 10^9}{T^{1.5}} * e^{\left[ \frac{-3430 K}{T} \right]} * [O_2] * [H_2]^{1.5}$	$\frac{mol H_2}{m^3 \cdot s}$
$r_9 = 6.4 * 10^7 * e^{\left[ \frac{57 \frac{kJ}{mol}}{RT} \right]} * [H_2]$	$\frac{mol H_2}{m^3 \cdot s}$
$r_{10} = 2.6 * 10^6 * e^{\left[ \frac{44 \frac{kJ}{mol}}{RT} \right]} * [H_2]$	$\frac{mol H_2}{m^3 \cdot s}$
$r_{11} = 480 * e^{\left[ \frac{106 \frac{kJ}{mol}}{RT} \right]} * [CH_4]^{0.5}$	$\frac{mol CH_4}{m^2 \cdot s}$
$r_{12} = 1.585 * 10^{10} * e^{\left[ \frac{200.841 \frac{kJ}{mol}}{RT} \right]} * [O_2]^{0.8} * [CH_4]^{0.7}$	$\frac{mol CH_4}{m^3 \cdot s}$

### S3 Methanation unit.

The methanation unit is modelled as a series of three adiabatic reactors with interstage cooling.

Each reactor is modeled by a 1D pseudo-homogeneous model. To describe axial temperature

and concentration profiles in each fixed-bed methanation reactor, a numerical 1D model has

been used. The material balances for each gas components (i=CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub>, H<sub>2</sub>O) was written as:

$$\varepsilon_g \frac{\partial c_i}{\partial t} = -u \frac{\partial c_i}{\partial \xi} - (1 - \varepsilon_g) \rho_c r_i \quad (\text{SI.24})$$

Where,  $\xi$ . is the dimensionless axial position belonging in [0,1],  $\varepsilon_g$  represent the bed porosity,  $c$  the gas concentration,  $\rho_c$  the packed-bed density and the rate of consumption or formation of i-species (i=CH<sub>4</sub>, H<sub>2</sub>, H<sub>2</sub>O, CO, CO<sub>2</sub>),  $r_i$ , is determined by summing up the reaction rates of that species in all the reactions  $R_j$  (see Table S5) according to the stoichiometric coefficient ( $v_{i,j}$ ) as follow:

$$r_i = \sum_{j=1}^2 v_{i,j} R_j \quad (\text{SI.25})$$

The gas superficial velocity ( $u$ ) has been calculated as follow:

$$u(\xi, t) = \frac{PM_{in}T(\xi, t)}{PMT_{in}} \quad (\text{SI.26})$$

where the subscript *in* represent the inlet conditions.

Finally, the energy balances were written as:

$$\left( c_{pg} \rho_g \varepsilon_g + c_{pc} \rho_c (1 - \varepsilon_g) \right) \frac{\partial T}{\partial t} = -u c_{pg} \rho_g \frac{\partial T}{\partial \xi} + \sum_{j=1}^3 (-\Delta H_{Rj}) r_j \rho_c \quad (\text{SI.27})$$

where  $\rho$  and  $c_p$  represents the density and heat capacity for gas (g) and solid (c), respectively, while  $\Delta H_{Rj}$  is the reaction enthalpy for  $j$  reaction.

The complete CO/CO<sub>2</sub> methanation reaction scheme is reported in. Table S5.

Table S5- Reactions scheme and associated standard enthalpies of reactions

Reaction	$\Delta H_{298} [kJ mol^{-1}]$	
$CO + H_2 \rightleftharpoons CH_4 + H_2O$	-206	R1
$CO_2 + H_2 \rightleftharpoons CO + H_2O$	41	R2
$CO_2 + 4H_2 \rightleftharpoons CH_4 + 2H_2O$	-165	R3

The kinetic rate models for syngas methanation over Ni-based catalyst can be expressed as:

CO methanation:

$$R1 = \frac{\frac{k_1}{P_{H2}^{2.5}} \left[ P_{CH4} \cdot P_{H2O} - \frac{P_{CO} \cdot P_{H2}^3}{K_{eq,1}} \right]}{DEN^2} \quad (S.28)$$

WGS:

$$R2 = \frac{\frac{k_2}{P_{H2}} \left[ P_{CO} \cdot P_{H2O} - \frac{P_{CO2} \cdot P_{H2}}{K_{eq,2}} \right]}{DEN^2} \quad (S.29)$$

CO<sub>2</sub> methanation:

$$R3 = \frac{\frac{k_3}{P_{H2}^{3.5}} \left[ P_{CH4} \cdot P_{H2O}^2 - \frac{P_{CO2} \cdot P_{H2}^4}{K_{eq,3}} \right]}{DEN^2} \quad (S.30)$$

$$DEN = 1 + K_{CH4} P_{CH4} + K_{CO} P_{CO} + K_{H2} P_{H2} + \frac{+K_{H2O} P_{H2O}}{P_{H2}} \quad (S.31)$$

$R_i$ ,  $K_{eq,i}$  and  $k_i$  are the reaction rate, equilibrium constant, and kinetic rate constant of reaction  $i$  ( $i = 1, 2, 3$ ), respectively, while  $p_j$  and  $K_j$  are the partial pressure and adsorption constant of species  $j$  ( $j = CH_4, CO, H_2, H_2O$ ), respectively. All the above kinetic parameters, depending on temperature, are given in Arrhenius-function form in Table S6.

Table S6 Kinetic parameters to calculate the reaction rates

$j$	$K_j$	$E_j$			$i$	$k_i$	$E_i$		
CH <sub>4</sub>	8.15e-4	bar <sup>-1</sup>	-38.28	kJmol <sup>-1</sup>	1	5.176e15	$\frac{kmol\ bar^{0.5}}{kgcat\ hr}$	240.1	kJmol <sup>-1</sup>
CO	10.08e-5	bar <sup>-1</sup>	-70.65	kJmol <sup>-1</sup>	2	2.395e6	$\frac{kmol\ bar^{-1}}{kgcat\ hr}$	67.13	kJmol <sup>-1</sup>
H <sub>2</sub>	7.5e-9		-82.9	kJmol <sup>-1</sup>	3	1.25e15	$\frac{kmol\ bar^{0.5}}{kgcat\ hr}$	243.9	kJmol <sup>-1</sup>
H <sub>2</sub> O	2.1745	bar <sup>-1</sup>	88.68	kJmol <sup>-1</sup>					

The mathematical model Eqs (SI.24)-(SI.27) has been completed with the following boundary and initial conditions:

$$\begin{aligned} c_i(0,t) &= c_{i,in}, c_i(\xi,0) = 0 \\ T(0,t) &= T_{i,in}, T(\xi,0) = T_0 \end{aligned} \quad (\text{SI.32})$$

Table S7 Operating condition of the system proposed.

<b>CLOU</b>	<b>Unit</b>	<b>Value</b>
Pressure	Pa	$1 \cdot 10^5$
Air-reactor temperature	K	923.15
Fuel reactor temperature	K	1173.15
<b>MU</b>		
first reactor temperature	K	553
first reactor pressure	Pa	$2 \cdot 10^6$
second reactor temperature	K	573
second reactor pressure	Pa	$2 \cdot 10^6$
third reactor temperature	K	593
third reactor pressure	Pa	$2 \cdot 10^6$
<b>PEM</b>		
outlet pressure	Pa	$4 \cdot 10^6$
H <sub>2</sub>	Nm <sup>3</sup> /h	100
electricity consumption	kWh/kgH <sub>2</sub>	46.6

Table S8 Parameters used to evaluate economic performance.

<b>Parameter</b>	<b>Unit</b>	<b>Value</b>
Specific cost of sludge conditioner	€/kg	1.40
Specific cost of electricity		
consumed	€/kWh·kg	$7.20 \cdot 10^{-2}$
produced	€/kWh·kg	$6.75 \cdot 10^{-2}$
Specific cost of oxygen carrier	€/kg	15.50
Specific cost of water	€/kg	$1.40 \cdot 10^{-3}$
Specific cost of methane	€/kg	$2.02 \cdot 10^{-1}$
Specific cost of Ni catalyst	€/kg	43.0
Specific cost of transportation	€/km·kg	$1.75 \cdot 10^{-5}$
Specific cost of landfill disposal	€/kg	0.5

Table S9 Technical performances of the proposed system.

	Unit	sewage sludge 1	sewage sludge 2
<b>Amount to be sent in landfill</b>	kg/h	0.31	0.24
<b>Generated thermal power</b>	kW/h	2.59	2.68
<b>Number of HP-PEM cell</b>	-		1
<b>CH<sub>4</sub></b>	kg/h		$7.37 \cdot 10^{-1}$
<b>H<sub>2</sub>:CH<sub>4</sub> residual ratio</b>	-		0.04

Table S10 Results as variation from the base line for the alternative sludge transportation distance, O.C. replacement ratio and PEM Energy consumption for sewage sludge 2.

	Sludge transportation			Oxygen carrier replacement ratio			Energy consumption PEM		
	Scenario			Scenario			Scenario		
	+5%	+10%	+25%	+5%	+10%	+25%	+5%	+10%	+25%
	<i>variation (from baseline)</i>								
Acidification terrestrial and freshwater	0%	0%	0%	15%	31%	77%	14%	28%	71%
Cancer human health effects	0%	0%	0%	0%	0%	1%	5%	10%	24%
Climate Change	0%	0%	0%	0%	1%	1%	1%	2%	6%
Ecotoxicity freshwater	0%	0%	0%	5%	11%	27%	1%	1%	4%
Eutrophication freshwater	0%	0%	0%	0%	0%	0%	0%	0%	0%
Eutrophication marine	0%	0%	0%	4%	8%	20%	12%	24%	59%
Eutrophication terrestrial	0%	0%	0%	4%	8%	21%	10%	21%	52%
Ionising radiation - human health	0%	0%	0%	1%	2%	5%	5%	9%	23%
Land Use	0%	0%	1%	1%	2%	5%	6%	12%	30%
Non-cancer human health effects	0%	0%	0%	1%	3%	7%	4%	7%	19%
Ozone depletion	0%	0%	0%	0%	0%	0%	0%	0%	0%
Photochemical ozone formation - human health	0%	0%	0%	1%	3%	7%	4%	7%	18%
Resource use, energy carriers	0%	0%	0%	0%	1%	1%	1%	2%	4%
Resource use, mineral and metals	0%	0%	0%	2%	4%	10%	3%	6%	15%
Respiratory inorganics	0%	0%	0%	1%	2%	6%	4%	8%	21%
Water scarcity	0%	0%	0%	1%	1%	3%	1%	2%	4%

Table S11 Comparison of different disposal cost and carbon cycle of alternative end of life strategies.

	Unit	Land spreading	Incineration	Landfill	Proposed system
C-EoL	€/kg	1,76E-01	2,48E-01	4,00E-01	1,80E+00
climate change	kg CO2eq/kg	3,26E-01	3,86E-01	3,57E+00	-2,18E+00
acidification	Mole of H+ eq./kg	-9,00E-01	3,23E+00	1,44E-03	1,01E-03
Photochemical Ozone Formation	kg NMVOC eq./kg	7,13E-02	1,65E+00	2,32E-03	-1,42E-03
Terrestrial Eutrophication	Mole of N eq./kg	9,94E+01	1,09E+01	4,60E-03	-1,66E-03
Freshwater Eutrophication	kg P eq./kg	3,60E+00	1,13E-01	9,65E-05	2,73E-02
Marine Eutrophication	kg N eq./kg	-2,25E-02	9,34E-01	9,81E-04	-1,34E-04



Table S12 Raw numerical results of the hot-spots analysis.

	Conventional scenario						Proposed scenario - SS1						Proposed scenario - SS2					
	Thermal energy credit	Drying	CLC	PEM	Transport	Waste in landfill	Thermal energy credit	Drying	CLC	PEM	Transport	Waste in landfill	Thermal energy credit	Drying	CLC	PEM	Transport	Waste in landfill
Water scarcity	-47.26%	40.30%	0.00%	0.00%	0.87%	11.58%	-34.20%	9.40%	3.92%	52.32%	0.16%	0.00%	-33.68%	9.40%	4.79%	51.78%	0.00%	0.44%
Respiratory inorganics	-9.75%	28.18%	0.00%	0.00%	31.41%	30.67%	-21.50%	15.97%	12.50%	49.07%	0.00%	0.96%	-20.63%	15.51%	15.07%	46.97%	0.00%	1.83%
Resource use - minerals and metals	-11.05%	85.42%	0.00%	0.00%	1.16%	2.37%	-3.66%	2.31%	38.19%	55.56%	0.00%	0.28%	-3.22%	2.08%	43.98%	50.44%	0.00%	0.27%
Resource use - energy carriers	-26.72%	17.36%	0.00%	0.00%	15.74%	40.18%	-78.94%	7.87%	2.55%	10.19%	0.00%	0.46%	-78.50%	7.64%	3.70%	9.72%	0.00%	0.43%
Photochemical ozone formation - human health	-6.27%	6.48%	0.00%	0.00%	3.01%	84.24%	-63.27%	7.41%	9.03%	19.44%	0.00%	0.85%	-61.97%	7.18%	11.11%	18.98%	0.00%	0.76%
Ozone depletion	0.00%	99.77%	0.00%	0.00%	0.23%	0.00%	-91.99%	6.85%	1.16%	0.00%	0.00%	0.00%	-91.56%	7.18%	0.95%	0.00%	0.00%	0.32%
Non-cancer human health effects	-4.09%	2.08%	0.00%	0.00%	3.70%	90.12%	-2.35%	0.23%	11.11%	71.76%	0.00%	14.55%	-1.91%	0.23%	12.96%	67.13%	0.00%	17.76%
Land use	-55.44%	9.03%	0.00%	0.00%	21.99%	13.54%	-26.28%	5.79%	6.48%	57.64%	0.70%	3.11%	-25.41%	5.56%	8.10%	56.02%	0.93%	3.98%
Ionising radiation - human health	-65.88%	9.03%	0.00%	0.00%	1.39%	23.70%	-23.67%	8.80%	9.49%	56.94%	0.00%	1.10%	-22.80%	8.56%	11.57%	55.09%	0.00%	1.97%
Eutrophication terrestrial	-10.62%	14.81%	0.00%	0.00%	6.02%	68.55%	-55.44%	8.56%	11.34%	23.84%	0.00%	0.81%	-54.57%	8.56%	12.96%	23.15%	0.00%	0.75%
Eutrophication marine	-5.40%	12.04%	0.00%	0.00%	2.55%	80.02%	-55.00%	11.34%	9.72%	23.61%	0.00%	0.32%	-53.70%	11.34%	11.11%	22.69%	0.00%	1.16%
Eutrophication freshwater	0.00%	13.89%	0.00%	0.00%	0.01%	86.10%	0.00%	0.02%	99.96%	0.00%	0.00%	0.02%	0.08%	0.00%	100.00%	0.00%	0.00%	-0.08%
Ecotoxicity freshwater	-14.53%	12.73%	0.00%	0.00%	11.57%	61.16%	-19.76%	4.86%	65.74%	9.26%	0.00%	0.38%	-16.71%	4.17%	70.60%	7.87%	0.00%	0.65%
Climate change	-4.09%	2.08%	0.00%	0.00%	2.31%	91.51%	-77.52%	7.41%	2.55%	12.50%	0.00%	0.02%	-76.76%	7.18%	3.70%	12.04%	0.00%	0.32%
Cancer human health effect	-12.79%	10.42%	0.00%	0.00%	7.18%	69.61%	-1.48%	0.23%	1.39%	95.37%	0.00%	1.53%	-1.48%	0.00%	2.10%	94.90%	0.00%	1.52%
Acidification terrestrial and freshwater	-11.49%	22.45%	0.00%	0.00%	5.32%	60.73%	-46.30%	8.80%	23.15%	21.07%	0.00%	0.69%	-43.69%	8.10%	27.32%	19.91%	0.00%	0.99%