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Technical–Economic Analyses of Electric Energy Generation by Biogas from Anaerobic Digestion of Sewage Sludge from an Aerobic Reactor with the Addition of Charcoal

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Abstract: This study aimed to obtain the energy recovery potential of the biogas produced from anaerobic digestion (AD) of the sludge from a wastewater treatment plant (WWTP), including the use of biochar as an additive for substrate co-digestion and catalyst for methane production. We carried out the following steps: chemical–physical laboratory analyses of sludge samples; the building, operation, and monitoring of an experimental prototype of a batch bioreactor of 2.5 L for the AD of the sludge (with and without the addition of charcoal); qualitative measurements of biogas; the study of charcoal morphology; and the projection of useful energy generation from the AD sludge after treatment. A study on the economic viability and avoided greenhouse gas (GHG) emissions was performed based on the experimental results. The substrate showed alterations in all the physicochemical parameters evaluated after AD, such as a reduction of 35% in the biochemical oxygen demand (BOD) analysis; the experiment carried out using biochar showed positive results regarding the speed of CH_4 production and a greater potential for energy recovery. Enterprises from 2000 kW onwards would present an internal rate of return (IRR) equal to or higher than the minimum attractiveness rate (MAR) of 15%. The USD 95.28/MWh tariff presented economic feasibility for the studied scenarios. WWTPs that produce enough sludge to generate power of 2000 kW would need to process the waste of 117,200 inhabitants with charcoal addition and 136,000 without charcoal. It would be possible to avoid the emission of 2307.97 tCO_2/year (2000 kW). According to the results obtained, this study revealed that using alternative energies based on anaerobic digestion and biochar can generate positive results regarding methane production, and its application as an energy source in a WWTP proved to be economically viable at a specific level of power production.

Keywords: biogas; sewage sludge; charcoal; renewable energy

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Citation: Garcia, C.R.; Hincapie, M.D.V.; Barros, R.M.; Córdova, M.E.H.; de Castro e Silva, H.L.; dos Santos, I.F.S.; Lora, E.E.S.; Filho, G.L.T.; Freitas, J.V.R.d.; Crispim, A.M.d.C.; et al. Technical–Economic Analyses of Electric Energy Generation by Biogas from Anaerobic Digestion of Sewage Sludge from an Aerobic Reactor with the Addition of Charcoal. *Methane* **2024**, *3*, 595–616. https://doi.org/10.3390/methane3040034

Academic Editor: Patrick Da Costa

Received: 13 September 2024
Revised: 21 October 2024
Accepted: 6 November 2024
Published: 2 December 2024

1. Introduction

In Brazil, according to the Brazilian Institute of Geography and Statistics (IBGE), Research Directorate, Department of Population and Social Indicators, National Survey of Basic Sanitation 2008—PNSB 2008, and data released by the Trata Brasil Institute [1], only 52.36% of the population has sewage collection, of which only 46% is treated. On 15 July 2020, the New Sanitation Framework was sanctioned through Law No. 14,026/2020 [2], which should bring and promote advances and attract new investments to universalize and qualify the provision of services in this sector. According to the National Water Agency (ANA), 27% of people do not have collection or treatment (without a sanitary collection service) [3]. The federal government aims to achieve universal access by 2033, ensuring that 99% of the Brazilian population has access to drinking water and 90% to sewage treatment and collection.

An alternative solution to the sanitation problem is anaerobic digestion (AD), an attractive process for treating organic waste that can control pollution and generate energy. Many agricultural and industrial wastes, including municipal solid waste (MSW) [4–6], are ideal for this process because they contain biodegradable materials. Anaerobic digestion can transform these wastes into renewable energy and products such as biopesticides, fertilizers, and bioplastics [6], reducing dependence on fossil fuels and greenhouse gas emissions [7,8].

The biogas produced by this process can generate electricity [9]. Furthermore, using carbon-based materials, such as charcoal, improves operational stability and increases biogas production by facilitating the transfer of electrons between bacteria, a process known as direct interspecies electron transfer (DIET) [10–13]. Charcoal has excellent porosity and a high adsorption capacity [10] and can be used as an additive for biomass co-digestion and to inhibit the production of acid gas H2S [12–15] (Hansen et al., 1999; Lü et al., 2016; Luo et al., 2015; Mumme et al., 2014), consequently increasing methane production and the potential for electricity generation [11,12,16–18].

The novelty of this study is the performance of experiments in batch reactors (2.5L) with sewage sludge supplied by the FANIA industry's wastewater treatment plant (WWTP) in Itajubá (Minas Gerais). In addition, this research investigated the use of co-digestion with biochar, based on a study by Shen et al. [16], to generate clean energy from biogas from the anaerobic digestion of sewage sludge. This study contributes to the UN Sustainable Development Goals (SDGs), specifically SDG 6 (Clean Water and Sanitation), SDG 7 (Affordable and Clean Energy), and SDG 11 (Sustainable Cities and Communities), by seeking to promote better sanitation conditions, affordable energy, and sustainability in cities.

Anaerobic Digestion

In general, organic matter is composed of particulates and water-insoluble polymers. The complex organic matter in the sludge used in biodigesters is a mass of biochemical compounds, such as carbohydrates, lipids, and proteins, that serve as a substrate for the metabolism of several microorganisms [19,20]. The genera of the most diverse bacteria are defined according to the degradation stages found in AD: hydrolytic, fermentative, acetogenic, methanogenic archaea, and sulfate-reducing bacteria (SRB) [20–22]. In the hydrolysis stage, hydrolytic enzymes are released by appropriate strains of hydrolytic bacteria which reduce complex compounds into simple molecules and dimeric substrates such as sugars, amino acids, and soluble fatty acids. Bacteria cause the breakdown of insoluble polymers into soluble monomers and oligomers [20]. This transforms carbohydrates into sugars, and lipids and proteins are converted to long-chain fatty acids and amino acids [20–23]. In the acidogenesis phase, bacteria consume these products and produce, among others, acetic acid, H_2, and mainly alcohol and CO_2 [20]. According to the authors, the acids produced are primarily acetic acid (CH_3COOH), propionic acid (CH_3CH_2COOH), and ethanol (C_2H_5OH). During acetogenesis, proton-reducing agents oxidize the longer volatile fatty acids (VFAs) and alcohols to acetic acid and hydrogen [20,24].

Consequently, the acetogenic and methanogenic stages depend on the products generated in the other stages. These develop a symbiotic system in which acetogenic bacteria produce hydrogen in their metabolism and methanogenic bacteria consume it, producing methane as the main final product [21–23]. In this context, hydrogenotrophic and acetoclastic methanogens are formed by reducing CO_2 with H_2 or cleaning cleaved acetic acid from the last-stage products: H_2, CO_2, and acetic acid [20].

A wide variety of inhibitory substances have been reported in anaerobic digestion. These substances are often also pointed out as the leading cause of failure in this process since they are present in substantial concentrations in sewage sludge (Kroeker et al. *apud* Chen et al. [4]).

The acetogenesis phase releases H_2 in large quantities and imposes toxic effects on the anaerobic microbes necessary for this process. Further, the high partial pressure of H_2 made the thermodynamic process unfeasible [25]. According to the authors [25], some efficient electron transmissions between acetogens and methanogens can use H_2 as an electron carrier, improving organic biodegradation and CH_4 yield. During methanogenesis, DIET revealed a greater electron transfer efficiency than interspecies hydrogen transfer (IHT) and interspecies format transfer (IFT) [25].

There is a competition between methane-producing archaea and sulfate-reducing bacteria (SRB) for the common substrates (H_2 and acetate) during AD of sulfate-rich wastewater [18]. In anaerobic reactors, sulfate is reduced to sulfide by the action of SRB [22,26–28]. Ammonia rates (NH_4^+) were evaluated by Lü et al. [13] at up to 7 g-N/L as an inhibitor in the biodigestion process by controlling the pH of the sample and the test to obtain total Kjeldahl nitrogen. The authors also studied this process by assessing hydrogen sulfide (H_2S) production and its concentrations through gas measurements. The authors concluded that improved methanogenesis was followed by the colonization of the biochar closely bound fractions by *Methanosarcina*, and the selection of suitable biochar particle sizes was imperative in enabling the initial colonization of microbial cells.

Biogas is a by-product of AD composed of 50 to 80% methane (CH_4) and $CO_{2,}$ which can also contain H_2S, H_2O(g), H_2, N_2, and O_2. Thus, it comprises different gases, but the most significant are methane and carbon dioxide [29].

According to the Intergovernmental Panel on Climate Change Report—IPCC, methane (CH_4) is a highly potent greenhouse gas (GHG) contributing significantly to climate change. Over 100 years, 1 ton of CH_4 will induce a warming effect equivalent to 25 tons of CO_2 [30]. The global average atmospheric carbon dioxide was 409.8 parts per million (ppm for short) 2019 [31]. Estimated anthropogenic global warming increases per decade by 0.2 °C (likely between 0.1°C and 0.3°C) because of past and ongoing emissions [32]. According to Fonseca [33], biogas' lower heating value (LHV) can vary from 5000 to 7000 kcal/m^3, reaching 12,000 kcal/m^3 if all carbon dioxide is eliminated from the mixture.

On the other hand, charcoal is a forest by-product resulting from the pyrolysis of wood, also known as the carbonization or dry distillation of wood. It is a destructive production method. In the carbonization process, the wood is heated in a closed environment, in the absence or with controlled amounts of oxygen, at temperatures above 300 °C, giving off water vapor, organic liquids, and non-condensable gases and leaving coal as a product [34].

Activated carbon is called "activated" because it is produced at high temperatures (ranging from 700–1200 °C) and is a substance with a high degree of porosity and selectivity for collecting impurities; it is usually used to promote methane production in waste. Swine, due to inhibitory sulfide uptake [12,14], like other toxic compounds, improves buffering capacity and supports the immobilization of anaerobic microflora [25,35], reducing the impact of organic shock load or accelerating methanogenesis in the onset of CD [8]. Activated carbon can also facilitate the electron transfer of interspecies microorganisms [36] present in sludge, as it is highly conductive [37], which can be beneficial for methanogenic systems [38]. Unfortunately, using activated carbon as a filler is challenging due to its high added value. However, biochar, a carbonaceous material similar to activated carbon with a greater surface area and lower cost, since it is produced at lower temperatures (<700 °C), is porous and biostable and can be an alternative for application as the substrate [8–32,34–40]. In other words, the gasifier's product resembles coal, which has properties similar to activated carbon [41].

Many studies have been carried out on carbon-based additives for wastewater degradation in AD. However, the degradation of the activated sludge system's sludge from a wastewater treatment plant (WWTP) is difficult. The novelty of this research lies in the study of the effects of adding bio-based carbons in improved DIET mechanisms on the performance of the AD of sludge, despite its difficulty, from a real WWTP. In this study, charcoal was used and evaluated as an additive in AD due to its similarity to those products, ease of purchase, and low cost for improving the AD of sludge from a WWTP of Fânia Industry, Itajubá, Brazil.

2. Materials and Method

The samples used in the AD process were collected from the Fânia Industry WWTP (reactor 1 of the activated sludge system) in Itajubá (Minas Gerais, Brazil). After collecting samples, they were stored in a refrigerated container and taken to a freezer. Experimental prototypes (biodigesters) were built according to Ribeiro et al. [39], Pin et al. [42], and Cañote et al. [43]. The samples were thawed and submitted in batches to a constant temperature of 35 °C (as done by Ribeiro et al. [39]) for the AD process.

The sludge collected from Fânia's WWTP comes from the activated sludge system for biological treatment by prolonged aeration in batches (mean treated flow of 58 m^3/day) of the effluents from the bathrooms and kitchen (internal restaurant) for approximately 250 employees of the industry. The restaurant can serve 600 employees per day.

2.1. Experimental Prototypes

For this work, six biodigesters were built on a laboratory scale based on Ribeiro et al. [39]. Two triplicates of the AD system were made, one containing charcoal for the support process. The acronyms E1 and E2 describe experiments 1 and 2, without and with charcoal added to the digesters, respectively. The acronym EU represents the term experimental unit. EU1, EU2, and EU3 are triplicates of E1, which had no added charcoal. Therefore, EU4, EU5, and EU6 are triplicates of E2, which had charcoal added to the samples for co-digestion.

RefPET bottles of 2.5 L (refillable PET or reusable PET) were used to build the experimental prototypes, as they are more resistant than typical bottles (Figure 1a). Using the same proportions as the study carried out by Ribeiro et al. [39], three-quarters of the reactor's total volume (1.875 L) was used, leaving one-quarter of the container for gas storage (gasometer).

After being filled with the substrate, the six bottles were put in a plastic box immersed in water, with a thermostat and a 60 W automatic heater set at 35 °C to control the temperature (Figure 1b).

Gas leakage from a biodigester is an undesirable factor. To reinforce the insulation, all regions that could allow the gas to leak were glued with commercial epoxy glue, thus ensuring that the reactor remained sealed.

2.2. Physicochemical Analyses

This study comprised experimental research on sludge based on the studies by Cañote et al. [43], Ribeiro et al. [39], and Pin et al. [42]. The analyses were performed according to the Standard Methods for Examination of Water and Wastewater (APHA) [44], namely the biochemical oxygen demand ($BOD_{5,20}$)—5210 B; chemical oxygen demand (COD)—5220 D; temperature—2559 B; total solids (TS)—2540 B; total fixed solids (TFS)—2540 E; total volatile solids (TVS)—2540 E; pH—4500-H + -B; total Kjeldahl nitrogen (TKN)—4500-Norg-C; and electrical conductivity (EC)—D1125-14.

The analysis and characterization of the substrate aimed to quantify the content of solids (total, volatile, and fixed), organic material, and the reductions in organic matter before and after AD.

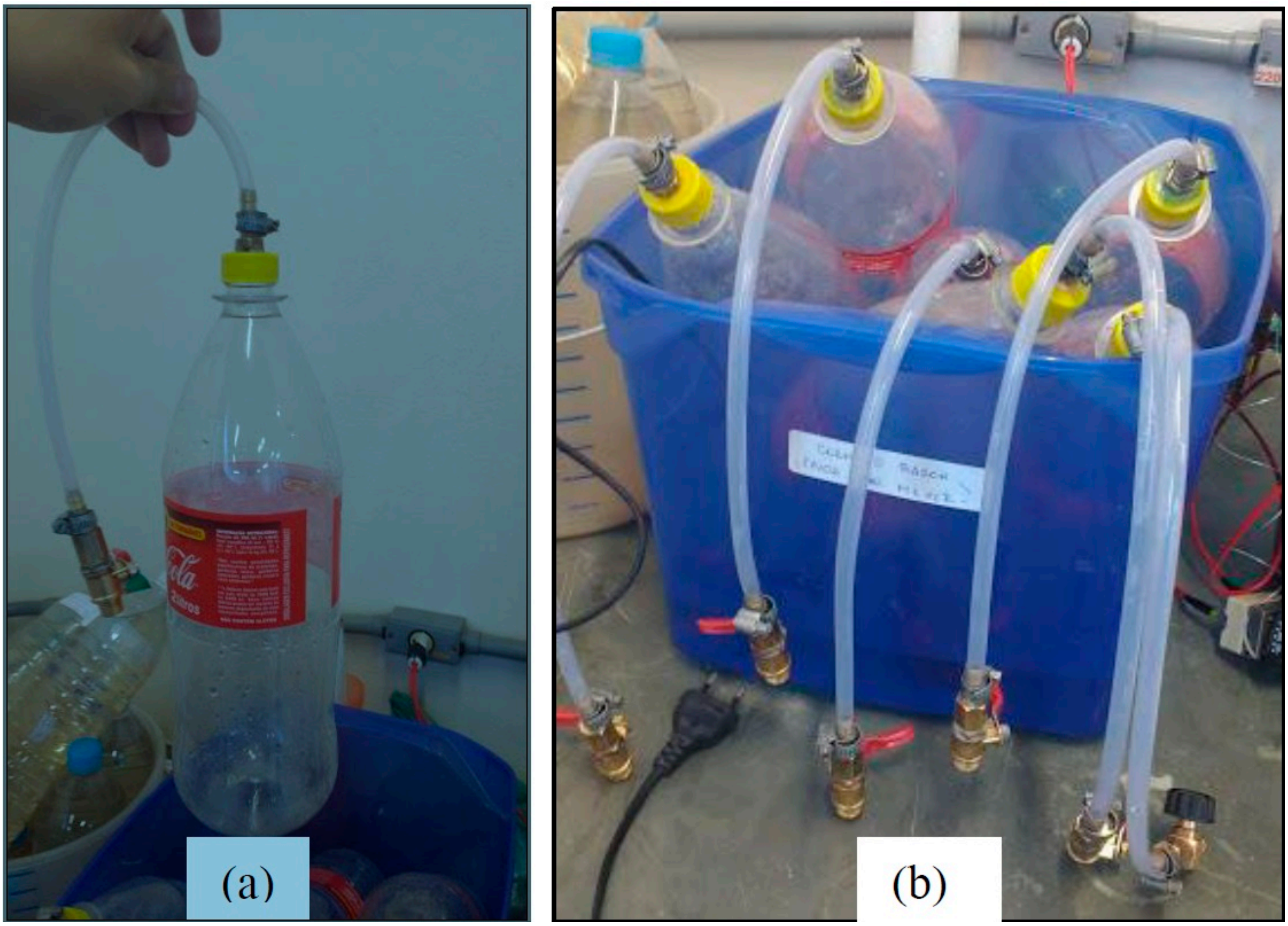


Figure 1. Experimental units: (**a**) bottle assembled (left); and (**b**) bottles immersed in the water system for heating (right).

2.3. Volume and Pressure Analyses

The measurements and volume corrections of the biogas produced followed the laws of Boyle and Gay-Lussac. A standard temperature and pressure (STP) of 1 atm and 20 °C were adopted to normalize the data. The pressure and meteorological conditions of the region of Itajubá (Minas Gerais—MG, Brazil) from April to June 2018 measured by the Meteorological Station of UNIFEI were considered, with a pressure of 0.92 atm and a mean temperature of 19.2 °C. The biogas temperature in the gasometer at the reading time was considered 35 °C. This value was the same temperature at which the experimental units were kept. The volume obtained was normalized by combining Boyle's and Gay-Lussac's laws. Equation (1) presents the phase equation of state. According to the STP, its unit is Nm^3/kg of CH_4 of volatile solid (VS).

$$\frac{Vo\ Po}{To} = \frac{V1\ P1}{T1} \tag{1}$$

where

Vo = corrected volume (m^3);

Po = corrected biogas pressure for 1 atm—1013.25 hPa;

To = corrected biogas temperature for 20 °C—293.15 K;

$V1$ = biogas volume in the gasometer (m^3);

$P1$ = pressure of the biogas plants at the time of measurement (hPa);

$T1$ = biogas temperature at the time of measurement in Kelvin (K).

2.4. Hydraulic Retention Time

Experimental data from biogas plants under different temperatures and hydraulic retention time (HRT) conditions were considered [45,46] to determine the HRT. Thus, at 35 °C, the ideal HRT was 25 days.

2.5. Biogas Measuring Equipment

Measurements were carried out using a gas analyzer (Landtec GEM5000—501944 series) with percentage measurements of CH_4, CO_2, O_2, H_2S, and CO gases available, in addition to the internal pressure of the reactor. For each measurement, the biogas outlet valve of the biodigester was closed and coupled to the gasometer sample/purge valve, which was later connected to the analyzer measurement port.

2.6. Available Power and Energy

Equations presented via software made available by the Environmental Sanitation Technology Company of the State of São Paulo [47] were used to determine the theoretical electrical power generation: Biogas® v. 1.0. The software Biogas® generation and energy use—effluents, version 1.0 is part of the products developed due to two agreements between the government of the State of São Paulo, through the Secretary of State for Environment and the Environmental Sanitation Technology Company (CETESB), and the Brazilian Federal Government, through the Ministry of Science and Technology: subsidies for the recovery and energy use of biogas generated in anaerobic wastewater treatment plants—ETAE (nº 01.0053.00/2001); and subsidies for the recovery and energy use of biogas generated at sites of solid waste disposal—LDRS (nº. 01.0054.00/2001).

The software models power and energy generation and analyzes the required investment, considering biogas production in the EUs' reactor under the STP for each indicator of organic matter present in the analyzed substrate. This study considered the production value of methane and biogas in volume (in m^3) per mass (kg) of the total solids (TS) of the sample. Values were calculated based on the measurements previously described. Equation (2) presents the power calculated from the biogas production.

$$P_u = \frac{Q_{bgd} \cdot PCI_{CH_4} \cdot E \cdot \eta \cdot 30}{t} \quad (2)$$

where

P_u = power, in kW;

PCI_{CH4} = the lower heating value of purified biogas in MJ/m^3 CH_4. (35.53×10^6 J/m^3CH_4; CETESB [47]);

E = methane system losses assumed to be 25%;

η = system efficiency (a Internal Combustion Engine, ICE), which was assigned a value of 33% (Bove and Lunghi [48]);

30 = assumption that a month has 30 days;

t = time that the system operated in a year (in seconds);

Q_{bgd} = amount of biogas produced in m^3/d, given by Equation (3). The values were multiplied by 30 days.

$$Q_{bgd} = \frac{30 \cdot \sum B_p \cdot Conc_i \cdot Q_{ti} \cdot M_{t_{bi}}}{VE} \quad (3)$$

where

Q_{bgd} = methane discharge (m^3/month);

30 = 30 days per month (day/month);

VE = specific volume of biogas (m^3);

B_p = biogas production ($kg_{biogas}/kg_{organic\ matter}$);

$Conc_i$ = methane concentration in percentage ($kg_{methane}/kg_{biogas}$);

Q_{ti} = number of effluents generating units;

M_t = total matter (kg).

Equation (4) is the basis for the supplied energy calculation, following Ribeiro et al. [39].

$$E = P_u \cdot CF \cdot 8760 \tag{4}$$

where

E = energy (MWh);

P_u = net power (MW);

CF = capacity factor (%) as 60% according to Ribeiro et al. [39];

8760 = number of hours in one year (24 h × 365 days), and is used to convert MW-year to MWh.

To be used in the Biogas® generation and energy use calculations in the CETESB software [47], the value was converted to kg of biogas per kg of TS, taking into account the biogas density, which is 5.70 kg/m^3, according to the website *Portal do Biogás*.

2.7. Structural Analyses of Charcoal

This study used commercial charcoal derived from reforested eucalyptus carbonized by pyrolysis. Mechanical grinding/spraying of the charcoal was carried out using a porcelain pistil to be used in the reactors. Qualitative, physicochemical, and structural analyses, such as scanning electron microscopy (SEM) using the Shimadzu brand device model SS-550 available at the Microscopy Laboratory of the Institute of Biomaterials (IFQ—UNIFEI), were carried out. SEM images of the sample relief were obtained using secondary-electron beams, which, among other features, showed porosity and topography. The amount of charcoal used in the triplicate followed what was proposed in the study by Shen et al. [16] concerning using biochar as an additive. The authors used 2.20 g of biochar for each gram of TS present in the substrate. Studies carried out using different types of charcoal with varying substrates in AD processes pointed out benefits such as the adsorption of inhibitory sulfide (H_2S) [12] and the acceleration of methanogenesis at the beginning of the activities of the biodigester [8]. This fact is possible due to the porosity of the coal, which has a relatively high surface area, thus allowing for the adhesion of methanogenic colonies [49].

2.8. Economic Studies

The economic and financial evaluation models used in this study are essential tools for assessing the economic viability of an enterprise. Among the several indexes, the Net Present Value Index (NPV), payback, and the Internal Rate of Return (IRR) can be used as the decision index, as it was considered in the present study. These models not only assess viability but also highlight potential economic benefits, providing an optimistic outlook on the study's implications.

Net Present Value (NPV)

To calculate the NPV, the results of the energy calculations and respective investment for electricity generation, such as revenues, by the sale of electricity (Equation (5)) and electricity generation were used. Equation (7) presents the calculation of the NPV based on a function $f(x)$ described by Equation (6). Finally, the NPV values were evaluated for the enterprise's first 20 years of implementation for each scenario.

$$Rec_{annual} = \left(E_{year} * V_s\right) - O\&M \tag{5}$$

$$f(x) = \frac{1}{\left[(1+i)^n\right]} \tag{6}$$

$$EVPL = FC_0 + \left[FC * f(x)_{acum}\right] \tag{7}$$

where

Rec_{annual}: annual revenue from the sale of energy (USD/kWh);

E_{year}: energy generated in one year (kWh/year);

V_s: Value of energy sold, with an assumed value of USD 95.28/kWh (BRL 509.74/kWh) de Silva et al. [50] and USD 72.91/kWh (BRL 390.00/MWh), according to the Brazilian Ministry of Mines and Energy (MME) ordinance no. 65/2018, as cited by UNICA [51]. The dollar value was consulted on the website of the Brazilian Central Bank [52]. This value is related to the day 27 November 2020, at BRL 5.3488 for each USD 1.00;

i: interest rate assumed to be 12% per year;

n: year evaluated;

$f(x)_{Acum}$: accumulated values of $f(x)$;

FC_0: Initial Cash Flow for electricity generation (USD);

FC : Expected cash flow over the years following the initial investment;

HR: total annual revenue (BRL);

$O\&M$: Operation and Maintenance is considered to be 5% of the investment, according to Brazil/Probiogas [53].

In addition, the attractiveness of the venture was analyzed. In the case of the non-financial viability of the project, a study was performed aiming to achieve the minimum population value that could become NPV positive, with a payback time inside a time of venture operation and Internal Rate of Return (IRR) exceeding or at least equal to the Minimum Attractiveness Rate (MAR) of 15.0%, as considered in Silva et al. [50]. Furthermore, the Internal Rate of Return (IRR) was also calculated based on relation to the NPV, i.e., when the NPV is null. The Equation (8) presents this relation.

$$\text{IRR} => \text{NPV} = FC_0 + \left[FC * \frac{1}{[(1+i)^n]} \right] = 0 \tag{8}$$

Investment values were consulted, taking into account the investment results of Silva et al. [50] in Figure 2 and Tolmasquim [54], both under Resolutions of the Brazilian National Electric Energy Agency (ANEEL) 482/2012 [55] and 687/2015 [56]. Equation (9) was extracted from the graph of Figure 2 and used in the present study.

$$Cost\ (USD) = 2431.90 * P + 158,074 \tag{9}$$

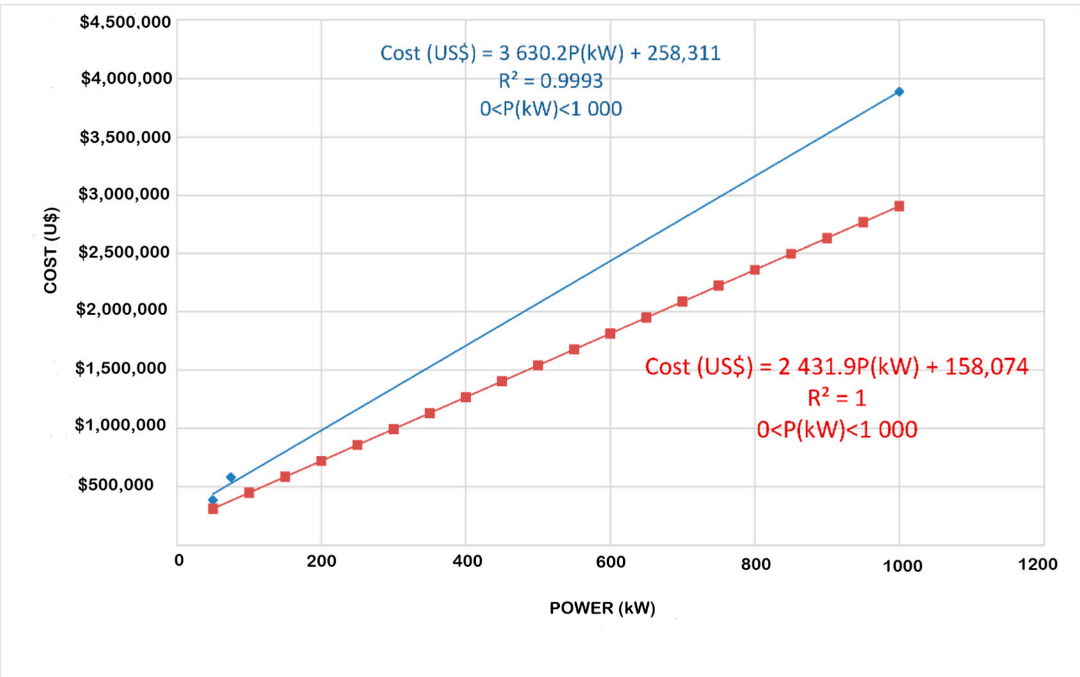


Figure 2. Values for investment in biogas thermal power plants. Source: Silva et al. [50].

According to studies conducted by the Energy Research Company (EPE, in Portuguese), the investment cost for electrical generation from anaerobic biodigesters with motor generators (internal combustion engines + generator set) is about USD 2402/kW [54] (Tolmasquim, 2016). The value of 5% per year of the investment cost can also be used for O&M regarding the anaerobic biodigestion plant with a motor-generator, according to Brazil/Probiogás [53] (2015) and also mentioned in Tolmasquim [54].

We used Silva et al. [50]'s value of BRL 509.74/kWh under ANEEL Resolutions 482/2012 [55] and 687/2015 [56]. Additionally, as mentioned, we considered MME ordinance nº 65/2018, as cited in UNICA [51], in which the new Specific Annual Reference Values (VRES, in Portuguese) were established for Distributed Generation Systems under Law nº 10,848/2004 [57].

Brazilian Law No. 13,203/2015 [58] established VREs. They represent the maximum amount distributors can pay for distributed generation and pass on to final consumers. VREs do not represent the final value to be passed on to the consumer, but the distributors organize the public call's ceiling price [59]. The authors mentioned that the price at which each distributor will effectively contract the distributed generation would result from this competitive process of public calls made by them. According to the EPE (undated), the distributed generation to which the VREs apply should not be confused with the distributed micro- and mini-generation regulated by the ANEEL mentioned above, Resolution nº 482/2012 [55]. The net energy metering system is currently applied in this modality, not involving the purchase and sale or VREs [59].

According to MME ordinance nº 65/2018 cited by UNICA [51], biomass and biogas have the following VRES (effective as of 1 March 2018):

- Residual Biomass—USD 65.24/MWh (BRL 349.00/MWh);
- Biogas—USD 72.91/MWh (BRL 390.00/MWh);
- Dedicated Biomass—USD 100.38/MWh (BRL 537.00/MWh).

Therefore, the economic viability was also simulated for the VRES value of USD 72.91/kWh (BRL 390.00/MWh), according to MME Resolution nº 65/2018 as cited by UNICA [51], four scenarios were produced, as follows:

- Scenario 1: A tariff of USD 95.28/MWh (BRL 509.74/MWh), according to Silva et al. [50], and investment according to Equation (9).
- Scenario 2: A tariff of USD 95.28/MWh (BRL 509.74/MWh), according to Silva et al. [50], and investment of USD 2 402/kW [54].
- Scenario 3: A VRES of USD 72.91/MWh (BRL 390.00/MWh), according to MME ordinance nº 65/2018 cited by UNICA [51], and investment according to Equation (9).
- Scenario 4: A VRES of USD 72.91/MWh (BRL 390.00/MWh), according to MME ordinance nº 65/2018 cited by UNICA [51] and investment of USD 2402/kW [54].

Economic feasibility values were calculated for various population sizes, ranging from 800 to 49,800 inhabitants. According to Tchobanoglous et al. [60], the volume of sludge produced per inhabitant was considered to be 0.6 (L/hab day).

2.9. GHG Emissions Avoided

Equation (10) calculates the annual GHG reductions (in tCO_{2eq}) using the methodology presented in Barros and Tiago Filho [61].

$$\begin{aligned} GHG_{avoided} &= \left(E_{factor_{NIS}} \cdot E_{generatyed_{system}}\right) \\ &- \left(E_{factor_{biogas}} \cdot E_{generatyed_{system}}\right) \end{aligned} \tag{10}$$

where

$GHG_{avoided}$: the annual reduction of GHG emissions from using an LFG/biodigester system ($tCO_{2eq/year}$);

$E_{factor_{NIS}}$: the baseline tCO_2 emission factor for the energy sector in 2019 (tCO_2eq/MWh), equal to 0.5181 tCO_{2eq}/MWh in 2019, according to the Brazilian Ministry of Science and Technology Brazil/MCT [62–64] (2020a; 2020b);

$E_{generatyed_{system}}$: the energy provided by the LFG/biodigester system per year (MWh/year);

$E_{factor_{biogas}}$: the tCO_2 emission factor (tCO_2eq/MWh), equal to 0.2986 tCO_{2eq}/MWh in 2019, according to Nielsen et al. [65].

We used the average emissions value (in tCO_{2eq}) to calculate the reduced GHG emissions as a function of generated electricity. According to the Brazilian Ministry of Science and Technology, the average value for electricity generation in the Brazilian National Interconnected System (NIS) was 0.5181 tCO_{2eq}/MWh in 2019 [62–64].

3. Results and Discussion

3.1. Biogas Composition in Reactors

This study performed ten readings (five for each experiment) using a GEM 5000 device. As shown in Table 1, data were obtained from each triplicate and their average values. The data measured by GEM 5000 were the percentages of CH_4, CO_2, O_2, CO ppm, and H_2S.

Table 1. Results of physicochemical analyses.

Parameter	Unit	Inlet	Outlet E1		Variation E1	Outlet E2		Variation E2
			Mean	Standard Deviation		Mean	Standard Deviation	
pH	-	6.3	8.6	0.1	37%	8.1	0.1	29%
Total solids (TS)	g/L	8.85	3.96	0.53	−55%	7.82	0.9	−12%
Fixed solids (FS)	g/L	3.26	1.26	0.18	−61%	2.91	0.26	−11%
Volatile solids (VS)	g/L	5.59	2.7	0.34	−52%	4.91	0.64	−12%
BOD	mg/L	2999	755.38	104.73	−75%	1071.45	92.52	−64%
COD	mg/L	1984.3	1168	6	−41%	1874.67	312.44	−6%
Total nitrogen	mg N-Nkt/L	112	438.47	30.62	291%	665.87	75.96	495%
Electrical conductivity	µs/cm	1276	1702.33	33.56	33%	1858.33	2.22	46%
COD/BOD	adimensional	0.66	1.55		134%	1.75		164%

One factor that should be considered when analyzing errors is the instability of working with microorganisms that behave differently, since simple environmental changes can significantly destabilize production. This fact can be observed well in each triplicate's values, which show standard deviation values close to each experiment's average, as indicated in Table 1. Figure 3 shows biogas production by the number of days of operation of the EUs.

It was noticed that the maximum production of CH_4 gas was anticipated and intensified at the beginning of the AD with the addition of charcoal when compared with the production without the additive. This result is similar to those obtained by Torri and Fabbri [18] and Zhao et al. [17]. This fact may be related to the buffering capacity of biochar. According to Chiappero et al. [66], the buffering capacity of biochar depends mainly on two factors:

A. Functional groups: During the anaerobic digestion (AD) process, volatile fatty acids (VFAs) rapidly accumulate, resulting in a medium with a low pH value. Some functional groups of biochar, such as amine, adsorb H+ and accept electrons, mitigating the sudden drop in pH.

B. Inorganic materials: The ash portion of biochar contains inorganic materials such as Ca, K, Mg, Na, Al, Fe, Si, and S. Alkali and alkaline earth metals (AAEMs) are responsible for biochar's alkalinity.

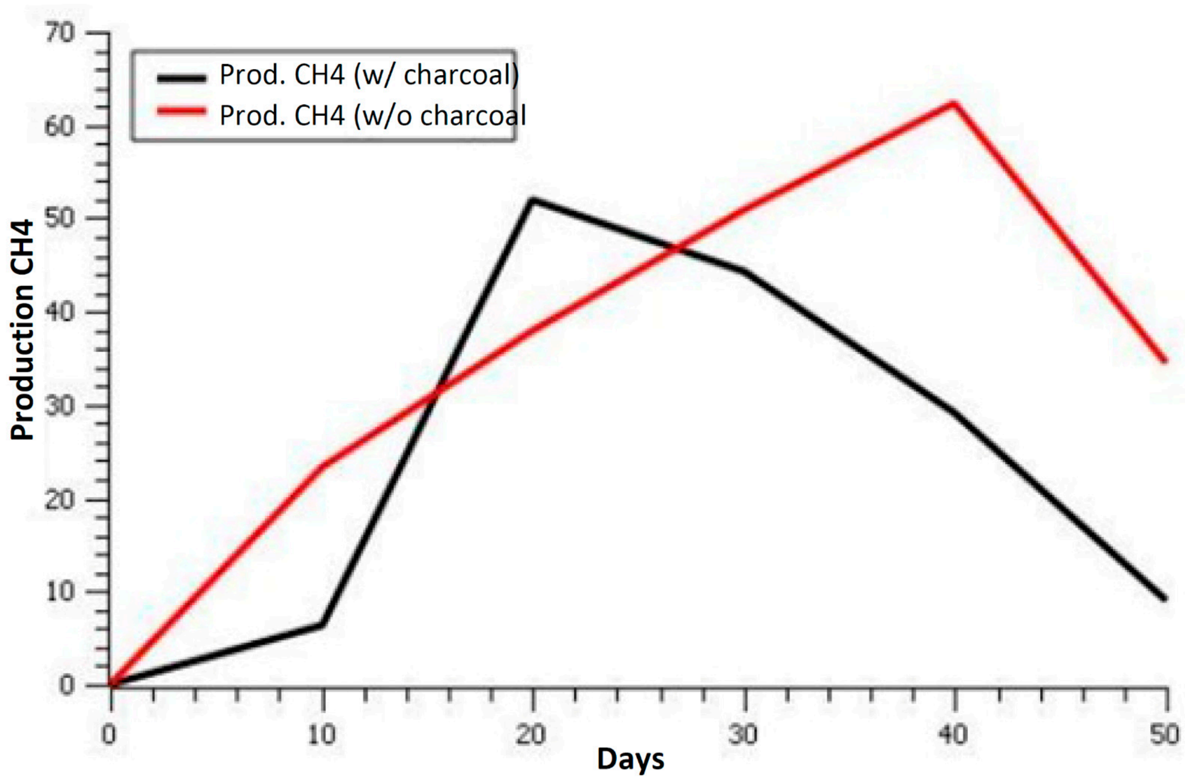


Figure 3. Comparative plot of the production of CH_4.

Therefore, the author summarizes that adding biochar can help to counteract the inhibition of VFA in the case of high loads of easily degradable wastes such as primary sludge. The alkaline nature of biochar, which determines its pH buffering capacity, may help to prevent VFA inhibition, and the ash fraction of biochar contains AAEMs, which may contribute to its acid buffering capacity, and the trace elements that are important for microorganisms [66,67]. In addition, porous biochar can support biofilm growth and protection for microorganisms, promoting the activity of microbial partners. This can enhance the degradation of syntrophic VFAs and methane production under high organic loads [66]. Furthermore, the biochar addition enhanced the cooperative oxidation of butyrate under high H_2 partial pressure [67].

Biochar can also reduce ammonia inhibition, leading to shorter lag phases and increased methane production compared to control reactors [66]. Therefore, biochar can help mitigate ammonia through direct means, such as cation exchange capacity, adsorption, and surface functionality, and indirect means, like DIET (Direct Interspecies Electron Transfer) and the immobilization of microorganisms. The specific impact of biochar depends on its characteristics, the digested substrate's properties, and the AD process's operational conditions, such as pH and temperature. According to Shao et al. [68], biochar may affect microorganisms through alternative pathways other than the DIET mechanism.

In a study by Cañote et al. [43], the most significant concentrations of methane (CH_4) in the biogas from the AD of ASS were 27.6% (in 8.56×10^{-4} m^3) and 41.9% (in 8.56×10^{-4} m^3). These results are not in compliance with those established by Resolution N° 685/2017 of the Brazilian Agency of Petroleum, Natural Gas and Biofuels [69], which determines the maximum concentration specifications for biomethane at 10 mg/m^3 H_2S (maximum); 3% CO_2 (maximum); 0.8% O_2 (maximum); 90% CH_4 (minimum); and 10% $N_2 + O_2 + CO_2$ (maximum).

Furthermore, as shown in Figure 4, the production of H_2S was reduced in both E1 and E2, while a reduction in hydrogen sulfide was seen only in E1.

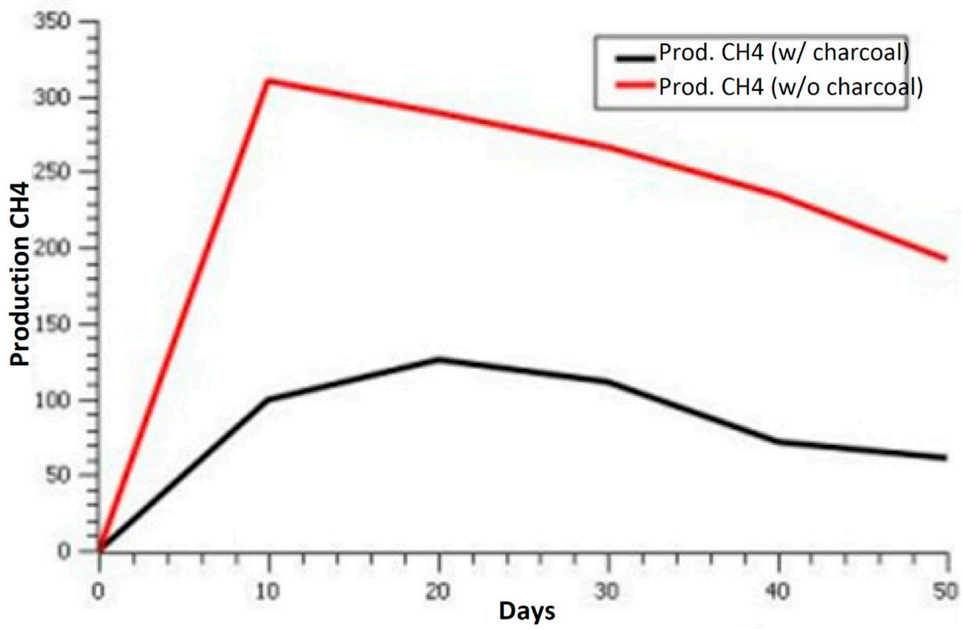


Figure 4. Comparative plot of the production of H_2S.

This control observed in the production of H_2S agrees with the studies of Lü et al. [13], Luo et al. [14], and Mumme et al. [15], in which the authors used co-digesters, such as biochar, to inhibit the production of H_2S. According to Xu et al. [70], the high alkalinity and abundant minerals in biochar are crucial for its strong H_2S sorption capacity and conversion of sulfur species. The authors preconized that it is noteworthy that the biochars produced are naturally alkaline and very effective in adsorbing H_2S without the need for alkaline pretreatments, which are typically used to boost the sorption capacity of activated carbons for H_2S.

Another factor observed is the presence of oxygen in the readings in all measurements, a gas that inhibits the action of methanogenesis, which occurs in anaerobic environments. However, this factor did not significantly interfere with the results obtained.

3.2. Physicochemical Results

Table 2 presents the results of the analyses carried out on the sludge before and after AD.

Table 2. Composition of the biogas produced.

Reading Date	Experiment/Experimental Unit		CH_4 (%)	Mean CH_4 (%)	CO_2 (%)	Mean CO_2 (%)	O_2 (%)	Mean O_2 (%)	CO (ppm)	Mean CO (ppm)	H_2S (ppm)	Mean H_2S (ppm)
19 April 2018	E2	EU1	15.4	23.3	14.8	21.1	12.5	9.5	2	13	113	310
		EU2	39.4		35.9		0.4		30		612	
		EU3	15.2		12.5		15.7		7		204	
	E1	EU4	8	6.4	6.8	6.4	17.4	17.8	4	10.3	223	99
		EU5	4.1		5.3		18.7		8		41	
		EU6	7.1		7.2		17.3		19		33	

Table 2. *Cont.*

Reading Date	Experiment/Experimental Unit		CH_4 (%)	Mean CH_4 (%)	CO_2 (%)	Mean CO_2 (%)	O_2 (%)	Mean O_2 (%)	CO (ppm)	Mean CO (ppm)	H_2S (ppm)	Mean H_2S (ppm)
25 April 2018	E2	EU1	34.7		6.9		17.3		2		307	
		EU2	49.4	38	23.3	20.8	11.9	14.5	6	5.3	267	289
		EU3	30.03		32.1		14.4		8		294	
	E1	EU4	57.5		23.1		14.6		5		142	
		EU5	43.2	51.9	24.5	26.3	5.7	6.9	13	13.7	102	126
		EU6	55		31.3		0.5		23		134	
4 May 2018	E2	EU1	43.5		24.7		11.8		0		116	
		EU2	59.1	50.8	31.4	29.1	9.5	10.1	2	1.7	197	267
		EU3	49.9		31.2		9		3		488	
	E1	EU4	45.1		20.8		12		2		132	
		EU5	38.8	44.2	7.1	19.2	17.7	14	1	2.7	48	111
		EU6	48.7		29.7		12.3		5		153	
15 May 2018	E2	EU1	58		30.4		3.2		3		178	
		EU2	64.2	62.2	22.8	24.7	8.4	10.2	3	2	149	234
		EU3	64.4		21		19		0		374	
	E1	EU4	24.3		3.2		20.6		0		41	
		EU5	32.4	29.1	26.6	15.8	15.6	18.7	1	1.3	149	72
		EU6	30.6		17.5		19.8		3		25	
21 May 2018	E2	EU1	16.6		11		19.3		0		321	
		EU2	51.3	34.5	26.6	19.7	2.1	13.7	3	1	190	192
		EU3	35.7		21.4		19.7		0		65	
	E1	EU4	7.2		5.7		17.6		0		105	
		EU5	13.9	8.9	9.1	6.4	18.7	18.5	0	0	72	61
		EU6	5.6		4.5		19.2		0		5	

It can be observed that the pH variation was lower in E2 (37%) than in E1 (29%), both having their most alkaline values after the experiment; respectively, 8.1 and 8.6. According to Chernicharo [71], the pH range between 6.0 and 8.0 is favorable for the growth of methane-producing microorganisms. Therefore, the environment of E2 would be more profitable for microbial development. Cañote et al. [43], when studying the AD of sludge from an activated sludge system (ASS), obtained variations of 7% (from 6.94 to 7.4) and 5% (from 6.80 to 7.12) in the pH values.

Moreover, the reduction in available organic matter was noteworthy. Regarding the VS parameter, the variation was −52% for E1 and −12% for E2. According to Johnravindar et al. [36], achieving improved methane production and a high VS removal efficiency is possible. The fixed solids (FS) in the digestate varied from 3.26 to 1.26 mg/L for E1 (a decrease of 61%) and from 3.26 to 2.91 mg/L for E2 (a reduction of 11%). The total solids (TS) showed variation in the digestate, ranging from 8.85 to 3.96 mg/L for E1 (a decrease of 61%) and from 8.85 to 7.82 mg/L for E2 (a reduction of 11%). According to Wang et al. [68], minerals and organic fractions are considered the fundamental components for constructing the buffer system. The authors performed a study in which TS (%) values were 88.7 ± 0.91 (vermicompost), 96.9 ± 1.03 (vermicompost biochar), 24.6 ± 0.93 (chicken manure), and 12.7 ± 0.70 (inoculum). The maximum daily biogas yield occurred during the first two days after inoculation, which was 22.6 and 29.1 mL/g_{ST} added for chicken manure digestion with vermicompost and chicken manure digestion with vermicompost biochar, respectively; however, CO_2 was the majority fraction. Wang et al. [68] described that the second biogas peak appeared on the 8th and 10th day for chicken manure digestion with vermicompost biochar and chicken manure digestion with vermicompost, respectively, and their corresponding CH4 yields were 12.4 mL/g_{TS} added and 7.98 mL/g_{TS} added, with CH_4 content greater than 60%. Considering the BOD parameter, it was lower in E2 (−64%, from 2999 to 1071.45 mg/L) when compared to E1 (−75%, from 2999 to 755.38 mg/l). Cañote et al. [43] obtained a variation of −59% (from 41.66 to 17.26 mg/l) and −19% (from 24.61 to 20.02 mg/L) for the BOD value, as well as −21% (from 85.67 to 68.00 mg/L) and −73% (from 198.00 to 48.00 mg/L) for the COD value. Factoring the COD parameter, the reduction in E1 was −41% (from 1984.3 to 1168.00 mg/L), while E2 was −6% (from 1984.3 to 1874.67 mg/L). The reduction in organic content presented by Felca et al. [72], who studied the AD of WWTP sludge, showed values similar to those given by Chernicharo [71] regarding COD and BOD, 40% and 70%, respectively. The values obtained by Felca et al. [72] demonstrated 44% to 75% reductions regarding TS, FS, and VS content. The initial COD/BOD ratio of 0.66 increased to 1.55 in E1 and 1.75 in E2.

The value found for total Kjeldahl nitrogen (TKN) increased 191% for E1 (from 112.0 to 438.47 mg/L) and 495% (from 112.0 to 665.87 mg/L) for E2. Cañote et al. [43] found that the TKN value increased by 25%, from 6.72 to 8.4mgN-Nkt/L. Johnravindar et al. [36] also investigated the soluble nitrogen released from protein degradation during AD by measuring the concentration of NH_4^+-N. The authors described that higher concentrations of NH_4^+-N were obtained from granular activated carbon (GAC) addition groups compared to the GAC-free group. Meanwhile, the maximum concentration of NH_4^+-N for all digestions was observed by Johnravindar et al. [36] in the first eight days (2020). The authors attributed this mainly to the highly degradable components in food waste studied by Johnravindar et al. [36]. Johnravindar et al. [36] related that more ammonia was produced in the GAC addition groups because of the relatively higher hydrolysis and acidification rates. Proteins were easily hydrolyzed under neutral pH conditions, leading to higher concentrations of NH_4^+ -N.

3.3. Biogas Production Using Charcoal

3.3.1. Charcoal Analyses

As suggested by Britto [34], qualitative analyses were carried out. When observing the noise of coal in contact with a surface, a metallic noise was observed, which means that the coal was hard, heavy, and of high density, factors that give it good quality and reflect an adequate manufacturing process.

Moreover, the charcoal was analyzed using a scanning electron microscope (SEM), Shimadzu model SS-550, so that the porosity of its structure could be seen. Figure 5a approximates the porous surface of the coal used in the reactors. Figure 5b approximates the surface inside the yellow rectangle in Figure 5a. Figure 6 shows the SEM detail of the charcoal porosity.

According to Wang et al. [49], charcoal (biochar) porosity is responsible for allocating methanogenic microorganisms, making them more stable and helping in biogas production. In the study of Johnravindar et al. [36], there was no microbial community on the GAC's porous carbon surface before anaerobic digestion, and the porous structure and irregular shape of the GAC surface provided spaces for the growth of microbial communities, which were dominant in biofilms. Johnravindar et al. [36] described that microbes were firmly attached to the GAC's surface as expected if the electrical connection was established between the cells and the GAC. The authors preconized that GAC's addition positively influenced volatile fatty acid (VFA) degradation and biofilm formation. This fact favors the essential syntrophic interaction for the degradation of acetate and propionate and bacteria growth [16,17,36]. Johnravindar et al. [36] justified that the electron shuttle (H_2 or formate) is no longer formed during acetogenesis. DIET exhibits more efficient electron transfer with conductive materials in the AD system than IHT [25]. Therefore, according to Johnravindar et al. [36], DIET could take advantage of its efficient electron transfer mechanism compared to mediated interspecies electron transfer. Such porosity can be observed in the charcoal structure used in our study, as shown in Figures 5b and 6. On the other hand, biochar in the digestate can be utilized as a fertilizer. Biochar-based fertilizers, which combine traditional fertilizers with biochar as a nutrient carrier, show promise in agronomy [73]. Biochar typically contains a small amount of essential nutrients, so it would be necessary to add substantial amounts of biochar to the soil, ranging from 10 to 50 tons per hectare, depending on the soil and biochar properties [73].

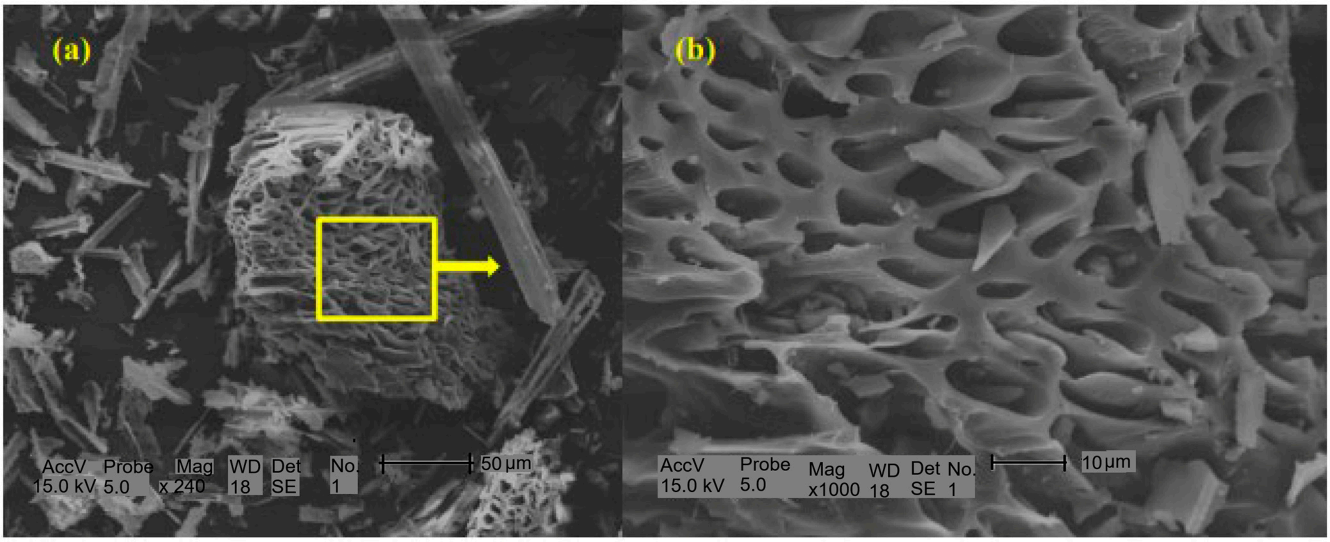


Figure 5. SEM of (**a**) charcoal and (**b**) SEM approximation of charcoal.

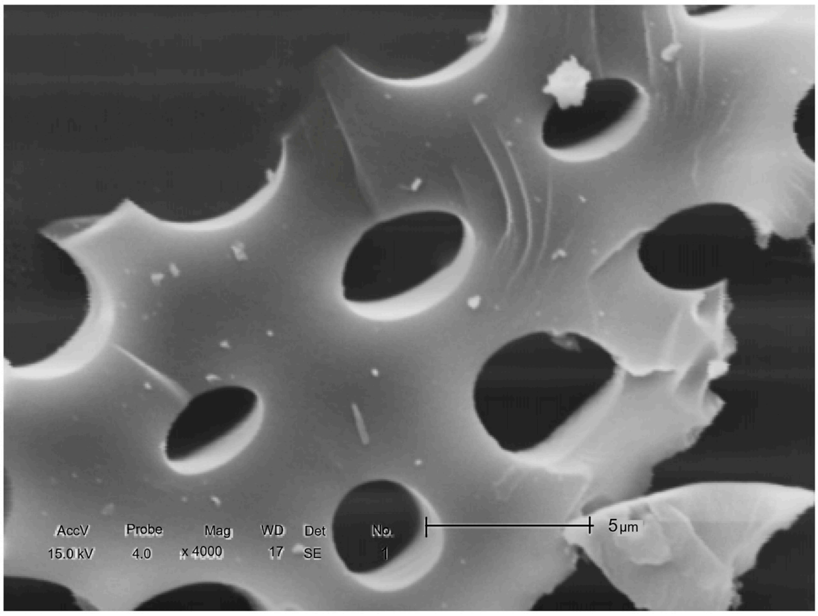


Figure 6. SEM detail of charcoal porosity.

3.3.2. Biogas Production Process Analysis

After the volume at STP and gas composition were tested in the laboratory, the methane yield from the substrate used on a laboratory scale was evaluated, which was 1.875 L. Table 3 shows the volume values in liters of biogas produced per day, liters of methane produced per liter of the substrate (sludge), and the volume (in Nm^3) of methane per m^3 of the substrate, in addition to presenting the methane yield (Nm^3 CH_4) concerning other essential parameters.

Table 3. Qualitative analyses of the biogas.

Sample	Q_{biogas} (L/day)	L_{CH4}/L_{sludge}	m^3 $_{CH4}/m^3$ $_{sludge}$	m^3 $_{CH4}/kg$ $_{substrate}$
E1	39.5617284	0.00023057	0.00000023	0.000000226
E2	38.19753083	0.00019877	0.0000002	0.000000195
Sample	m^3 $_{CH4}/kg$ $_{COD}$	m^3 $_{CH4}/kg$ $_{BOD}$	m^3 $_{CH4}/kg$ $_{TS}$	m^3 $_{CH4}/kg$ $_{VS}$
E1	0.0002825	0.0001028	0.0000472	0.0000798
E2	0.0018131	0.0001031	0.000193	0.0002923

The calculations were made using the results of the physicochemical analyses (variations) in both experiments. E2 demonstrated results from the physicochemical point of view due to its smaller reduction in these parameters (2923 $\times$ 10^{-4} Nm^3/kg of VS of CH_4). Conversely, E1 yielded more favorable results in production and enhanced effluent treatment due to reduced organic pollution (7.98 $\times$ 10^{-5} Nm^3/kg of VS of CH_4. The reduction values of the physicochemical parameters can be seen in Table 3. Cañote et al. [43] obtained from the ASS sludge samples yields of 1.9 $\times$ 10^{-3} Nm^3/kg of VS of CH_4 (1.9 Nm^3/t of VS of CH_4) with 50.45% VS and 9.7 $\times$ 10^{-3} Nm^3/kg of VS of CH_4 (9.7 Nm^3/t of VS of CH_4) with 17.57% of VS. Felca et al. [72] found values of 0.34 $\times$ 10^{-3} Nm^3/kg of VS of CH_4; 0.37 $\times$ 10^{-3} Nm^3/kg of VS of CH_4; 0.76 $\times$ 10^{-3} Nm^3/kg of VS of CH_4; and 17.31 $\times$ 10^{-3} Nm^3/kg of VS of CH_4.

3.4. Economic Study

Figure 7a–d present the NPV values regarding the three scenarios studied.

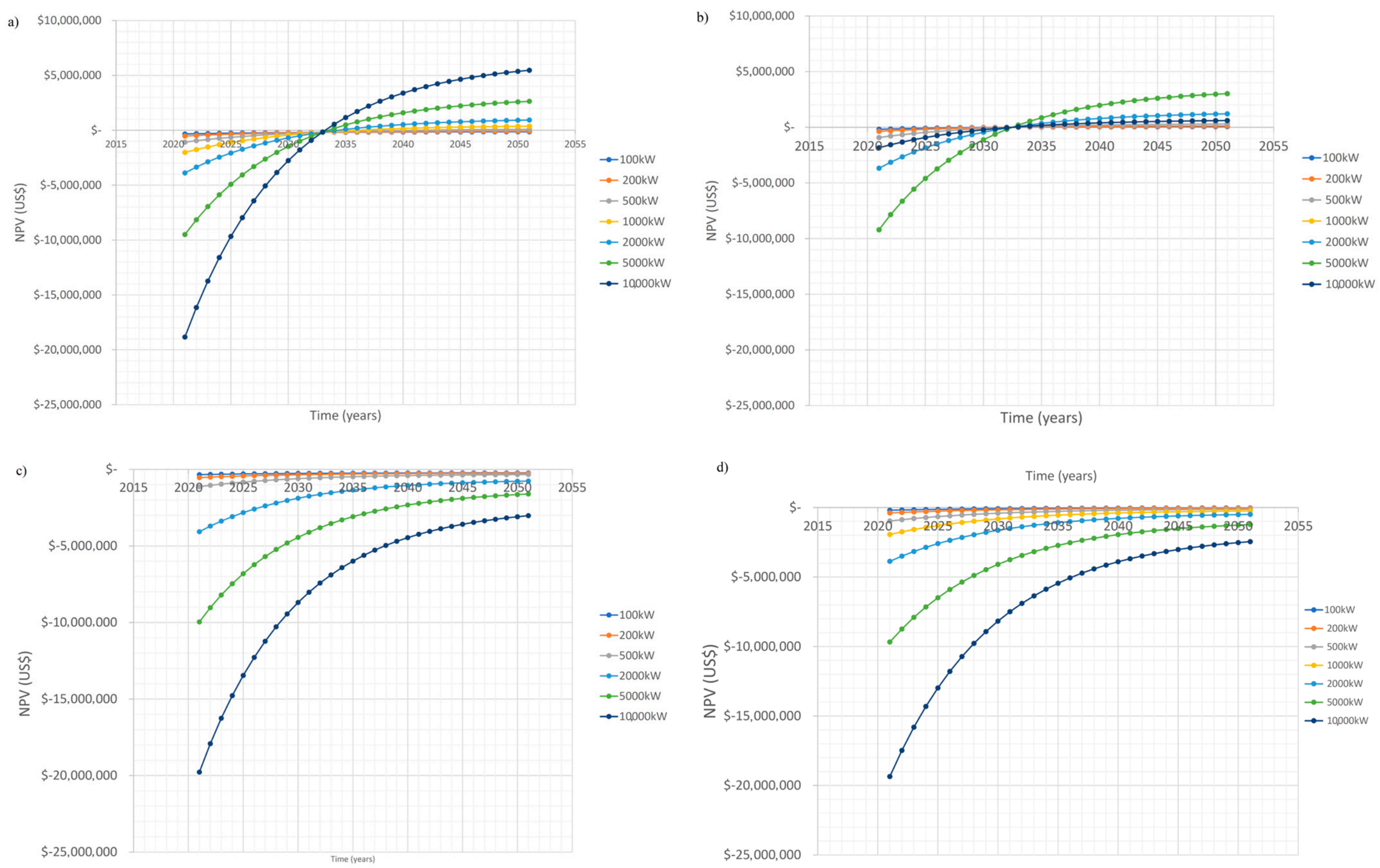


Figure 7. NPV (**a**) Scenario 1 (left superior); (**b**) Scenario 2 (right superior); (**c**) Scenario 3 (left inferior); (**d**) (right inferior).

According to Figure 7a,b, it is possible to verify that Scenarios 1 and 2 present economic feasibility. Regarding the power of 100 kW and 200 kW, there is no payback for Scenario 1. This scenario shows the payback for projects starting at 500 kW (in the 21st year). In Scenario 1, and enterprises with power of 1000 kW, the payback occurs in the 16th year, for 2000 kW in the 15th year, and for 5000 kW and 10,000 kW, both happen in the 14th year. As Scenario 2 was based on a cost of USD 2420.00/kWh, e.g., a constant incremental value for each energy unit, the payback value presented mathematically the same value for all the power values studied. The payback for all these amounts resulted in the 13th year. Scenarios 3 and 4 did not show economic viability. Figure 8 shows the IRR regarding Scenario 1 and Scenario 2 related to the minimum attractiveness rate.

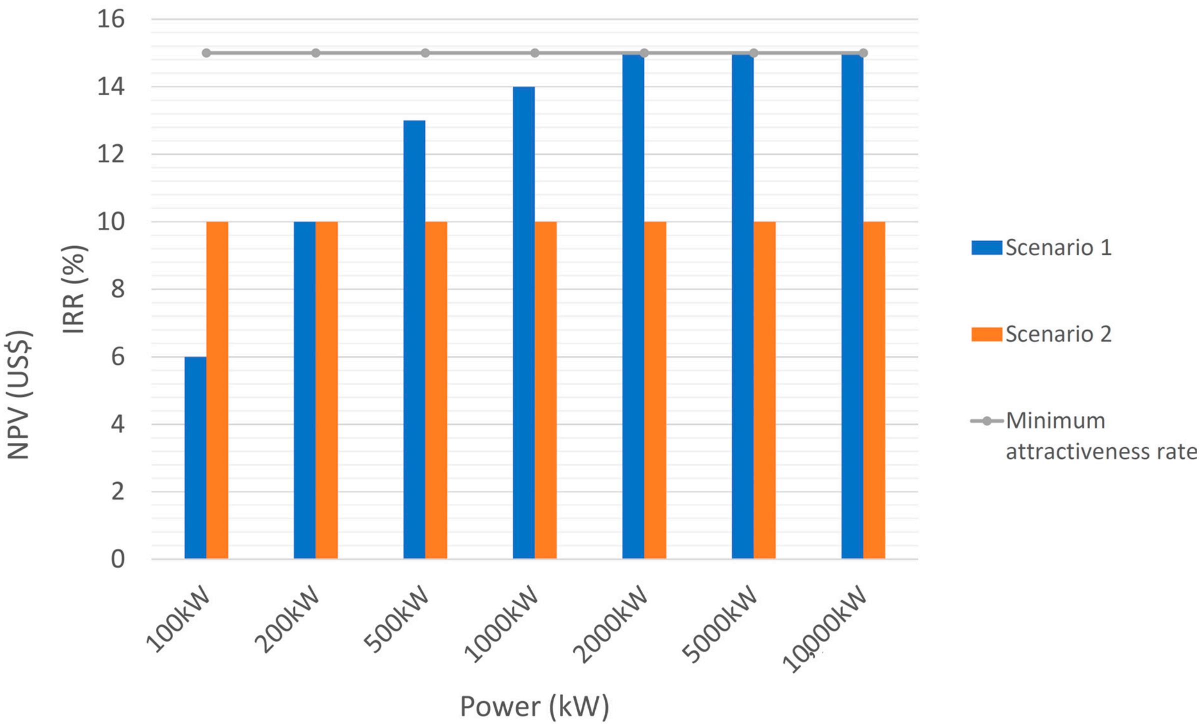


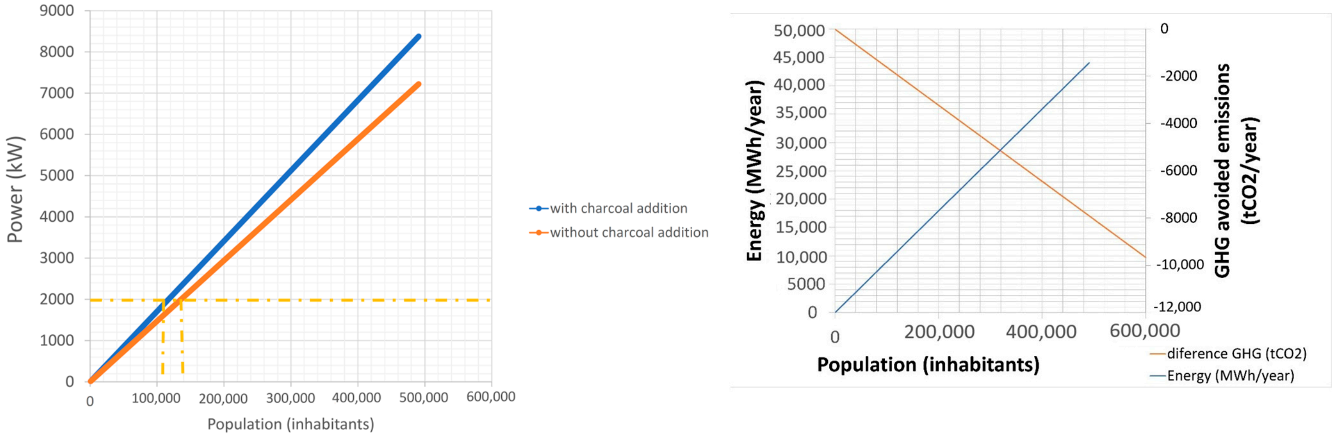
Figure 8. IRR regarding Scenario 1 and Scenario 2 related to minimum attractiveness rate.

According to Figure 8, the enterprises that showed payback within the studied operation time were only Scenarios 1 and 2. Scenario 1 presented an IRR of 6% (100 kW), 10% (200 kW), 13% (500 kW), 14% (1000 kW), and 15% for 2000 kW, 5000 kW, and 10,000 kW. Enterprises from 2000 kW onwards would have an IRR equal to or higher than the minimum attractiveness rate of 15%. Scenario 2 presented the same IRR values (10%) for all the studied power values.

3.5. Power, Energy, and Avoided GHG Emissions

Figure 9a presents the population versus Scenarios 1 and 2, and Figure 9b shows the energy generated and avoided GHG emissions for these scenarios.

Figure 9a shows the population contributing to WWTP that produces an amount of sludge at the end of the treatment, which results in the economic viability of the enterprises to generate electricity from biogas from the anaerobic digestion of this sludge. Enterprises from 2000 kW onwards had a payback within the plant's operation time and with an IRR value greater than or equal to the minimum attractiveness rate of 15%. In Figure 9a, it is possible to verify that the population contributing to the WWTP that produces enough sludge to generate 2000 kW of power is 117,200 inhabitants (with charcoal addition) and 136,000 (without charcoal). From an energy capacity of 10,000 MWh/year, considering the charcoal addition, it is possible to avoid the emission of 2307.97 tCO_2/year, as can be observed in Figure 9b, with incremental values of annual energy obtained and avoided emissions per year, according to power values addition.



(**a**) (**b**)

Figure 9. Population: (**a**) versus Scenario 1 and Scenario 2 (right); (**b**) energy generated and avoided GHG emissions versus Scenario 1 and Scenario 2 related to minimum attractiveness rate.

4. Conclusions

This study demonstrated the importance of public policies that encourage alternative forms of energy, such as the generation of biogas from sewage sludge using anaerobic biodigestion. An experimental reactor was built and successfully operated, allowing the monitoring of biogas production and the estimation of energy convertible into electricity. The addition of charcoal proved beneficial in improving the performance of anaerobic digestion (AD), increasing the removal of organic matter (COD) and the production of biogas.

The main results include the following:

- The pH variation was smaller in E2 (37%) than in E1 (29%), both reaching alkaline values (8.6 for E1 and 8.1 for E2).
- There was a reduction in organic matter: E1 presented a −75% reduction in BOD, while E2 reduced −64%. E1 obtained a −41% reduction for COD, and E2 −6%.
- The production of CH_4 was faster with the addition of charcoal, and the production of H_2S was interrupted in E1.
- Although the industrial results were low (0.19 kW and 0.79 kW for E1 and E2), E2 showed more significant potential for energy recovery.
- In the economic scenario, companies with power above 2000 kW had a positive return (IRR of 15% in Scenario 1 and 10% in Scenario 2).
- With the addition of coal, the emission of 2307.97 tCO_2/year can be avoided for plants generating 2000 kW.

Author Contributions: Conceptualization, R.M.B., G.L.T.F. and E.E.S.L.; methodology, R.M.B., G.L.T.F. and E.E.S.L.; software, R.M.B. and I.F.S.d.S.; validation, C.R.G., M.D.V.H., M.E.H.C. and H.L.d.C.e.S.; formal analysis, R.M.B. and J.V.R.d.F.; investigation, M.D.V.H., M.E.H.C. and H.L.d.C.e.S. and J.V.R.d.F.; resources, R.M.B. and C.R.G.; data curation, C.R.G. and M.D.V.H.; writing—original draft preparation, R.M.B. and C.R.G.; writing—review and editing, R.M.B., A.M.d.C.C. and A.J.M.d.O.P.; visualization, A.M.d.C.C. and A.J.M.d.O.P.; supervision, R.M.B.; project administration, R.M.B.; funding acquisition, R.M.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Minas Gerais State Research Support Foundation (*Fundação de Amparo a Pesquisa do Estado de Minas Gerais*—FAPEMIG) by granting a Scientific Initiation scholarship to Cornélio Ribeiro Garcia.

Data Availability Statement: The raw data supporting the conclusions of this article will be made available by the authors on request.

Acknowledgments: We thank the Coordination for the Improvement of Higher Education Personnel (CAPES) for the support given by the doctoral scholarship granted to Ivan Felipe Silva dos Santos and master's scholarships to Hellen Luisa de Castro e Silva and Maxi Estefany Huamán Córdoba. The authors would like to thank the National Council for Scientific and Technological Development (CNPq) for granting a productivity in research scholarship to Regina Mambeli Barros (P1D Process Number 303036/2021-4), to Geraldo Lúcio Tiago Filho, and Electo Eduardo Silva Lora (P1A). We would also like to thank the Minas Gerais State Agency for Research and Development (FAPEMIG) for granting the scientific initiation scholarship to Cornélio Ribeiro Garcia and for granting financial support for the "Improvement of biogas energy potential from anaerobic (co)digestion of solid organic waste as an incentive to renewable energy sources: substrate pre-treatment and co(digestion) aiming at Hydrogen use" (Process N.: APQ-00568-21) and for project RED-00090-21, "Theoretical-experimental evaluation of the production and use of green hydrogen in Minas Gerais." We thank FAPEMIG for granting the Doctorate scholarship (finance code I) to Adriele Maria de Cássia Crispim and the Master of Science scholarship to Aylla Joani Mendonça de Oliveira Pontes by FAPEMIG.

Conflicts of Interest: The authors declare no conflicts of interest.

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