JUSTIFICATION OF COMPONENT COMPOSITION OF SOLID COMPOSITE BIOFUEL BASED ON SEWAGE SLUDGE AND CEREAL STRAW

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Abstract. The disposal of sewage sludge is a pressing issue for municipal utilities, where the implementation of high-cost thermal drying technologies is economically unviable. Previous research by the authors has demonstrated the effectiveness of “sewage sludge - cereal straw” mixtures for aerobic composting (bioconversion). The aim of this work is to provide a theoretical justification for the use of such composite mixtures in the production of solid biofuel (fuel pellets), whilst simultaneously establishing a standardised Life Cycle Inventory (LCI) for subsequent environmental assessment. The input data for the modelling were the results of laboratory studies of the physical and chemical properties of sludge from the Boiarka wastewater treatment plant (average moisture content – 75.41%, carbon and nitrogen content in dry matter – 24.04% and 3.63%) and the characteristics of wheat straw (moisture content 12%, net calorific value of dry matter 17.6 MJ·kg^{-1}). All mass and energy balances were normalised to a single functional unit – 1 kg of the finished composite mixture. Calculations showed that adding up to 60% straw to sludge allows the moisture content of the mixture to be reduced to 31-39% without the use of additional thermal energy. At this moisture content, according to the modelling results, the lower calorific value of 1 kg of the resulting pellets will be 9.78-10.32 MJ·kg^{-1}. Analysis of the chemical composition demonstrates that “diluting” sludge with biomass further reduces the concentration of heavy metals in the ash residue to a safe level. The mass and energy parameters obtained confirm the energy efficiency of the co-pelletisation technology and form the basis for further multi-criteria life cycle assessment (LCA) of alternative utilisation scenarios.

Keywords: sewage sludge, composite biofuel, co-pelletisation, mathematical modelling, life cycle inventory.

Introduction

The increasing volume of sewage sludge and the search for effective technologies for its utilisation represent one of the most significant and pressing environmental and engineering challenges. Traditional methods of managing sludge, such as accumulation in sludge storage areas or disposal in landfills, lead to secondary contamination of soil and water bodies, as well as greenhouse gas emissions [1]. In accordance with modern principles of the circular economy, sludge is regarded not as waste but as a valuable resource containing a significant amount of organic matter and biogenic elements [2].

The current regulatory framework of the European Union, in particular the Council Directive 86/278/EEC [3], encourages the use of sewage sludge as an organic fertiliser, whilst preventing its harmful effects on soil, vegetation, animals and human health. This directive sets strict maximum permissible concentrations of heavy metals both in the sludge itself and in the soil into which it is applied; however, it does not contain requirements regarding a pollutant such as microplastics.

It is known [4] that microplastics accumulate in high concentrations in sewage sludge; after the sludge is disposed of, microplastics enter the environment, leading to their spread and posing risks to the environment and human health. The issue of microplastics and the absence of relevant provisions in EU Council Directive 86/278/EEC is leading to a shift in the regulatory policies of EU countries. For instance, the German Sewage Sludge Ordinance prohibits the use of sludge on fields [5]. Instead of using it as fertiliser, the regulation provides for the thermal treatment of sludge with mandatory phosphorus removal (from 2029-2032) for its subsequent safe use as a pure mineral fertiliser.

Currently, one of the most promising areas for sewage sludge utilisation is its use for energy generation. Due to its high organic content (up to 70-80% on a dry weight basis), the calorific value of dried sludge can be comparable to that of low-grade coal or peat [6]. However, the main technological obstacle to the direct use of sludge as fuel is its high moisture content after mechanical dewatering, which is usually 75-85%. Pre-drying the sludge to the required moisture content is an extremely energy-intensive process. The energy costs of evaporating water often exceed the energy value of the resulting fuel, making such single-technology approaches economically unviable in the context of the modern energy market [7]. An effective engineering alternative is the production of solid composite biofuel through co-pelletisation. The method involves mixing wet sludge with dry biomass, in particular agricultural waste. Ukraine has a significant surplus of cereal straw, which is characterised by low moisture content (10-15%), high calorific value and is suitable for use in biofuel technologies [8]. The

DOI: 10.22616/ERDev.2026.25.TF137

use of cereal straw allows the moisture content of the raw material mixture to be optimised through “passive drying” (absorption of free moisture by dry biomass), completely avoid the costly thermal drying stage, and at the same time improve the physical, mechanical and thermal properties of the finished pellets, as well as reduce emissions of harmful substances during combustion [9; 10].

In our previous studies [11], we proposed the effective use of mixtures of “sewage sludge + cereal straw” for aerobic composting (bioconversion) processes to produce organic fertilisers. In addition, a technology for the production of composite fuel pellets based on other municipal waste (sewage sludge and fallen leaves) was tested [12]. The choice between biological (composting) and thermochemical (incineration) disposal of municipal solid waste requires a comprehensive environmental assessment using the Life Cycle Assessment methodology. However, for a correct comparative LCA analysis of both technologies, it is critically important to have a single, technologically sound reference point – an optimised base mixture of components. Thus, determining the optimal physico-chemical parameters of the composite is a necessary step in establishing a reliable LCA for further environmental studies.

Consequently, the aim of this work is to provide a theoretical justification for the component composition of a composite mixture based on sewage sludge and cereal straw to ensure the energy efficiency of the solid biofuel pelletisation process and to establish a standardised inventory database for life cycle assessment.

Materials and methods

The research methodology is based on a combination of laboratory analysis of the physico-chemical properties of the raw materials and the conduct of a factorial numerical experiment to optimise the composition of the composite biofuel. With a view to the subsequent use of the results obtained for a comparative environmental LCA analysis of waste utilisation technologies, all material and heat balance calculations were normalised to a single functional unit (FU) – 1 kg of the finished composite mixture based on sewage sludge and cereal straw – wheat. This allowed for the standardisation of raw material input streams and the output parameters of the finished product.

The main subject of the study was sewage sludge collected at the operational wastewater treatment plant of the municipal enterprise in Boiarka (Kyiv region, Ukraine). This facility is a typical example of a treatment plant in a small town, where domestic wastewater accounts for over 95% of the inflow, thereby minimising the risk of critical contamination by industrial pollutants. Samples were taken directly from open sludge storage areas (Fig. 1, a) in accordance with the guidelines of ISO 5667-13:2011 [13]. An improved grain sampling probe (Fig. 1, b) was used for sampling, which allowed the compacted and vegetation-covered top layer of the storage site to be effectively penetrated and samples to be taken from depths of up to 3 m.

To ensure the composite sample was as representative as possible and to account for the uneven natural drying of the sludge in the different storage areas (a total of three equally sized rectangular areas, with the sludge feed point located at the centre of the shorter side), spot samples were taken using the modified “envelope” method. Six samples were taken from each of the three sections, with three samples taken along the longer sides of each section at a distance of one quarter of the length of the shorter side from the edge of the section. Spot samples were taken from surface and deep layers (at a depth of up to 2 m), after which they were mixed to average the physico-chemical parameters before being transported to the laboratory in plastic containers.

The physico-chemical parameters of the collected sewage sludge samples were determined at the Ukrainian Laboratory for the Quality and Safety of Agricultural Products, accredited in accordance with the requirements of DSTU EN ISO/IEC 17025:2019 (certificate No. 20724), using standard methods. Moisture and dry matter content were determined by the gravimetric method in accordance with ISO 11465:1993 and EN 12880:2000; organic matter content – in accordance with DSTU 8454:2015; total nitrogen content – in accordance with DSTU 7911:2015; pH determination – in accordance with DSTU 7882:2015; cadmium content – in accordance with EN 13346:2000 and the specialised working method RM.UL.5.4-90 [14].

Based on the results of the analyses, the following baseline parameters of the sewage sludge were established for modelling (average values): moisture content – 75.41%, organic matter (carbon) content – 24.04%, nitrogen content in dry matter – 3.63%.

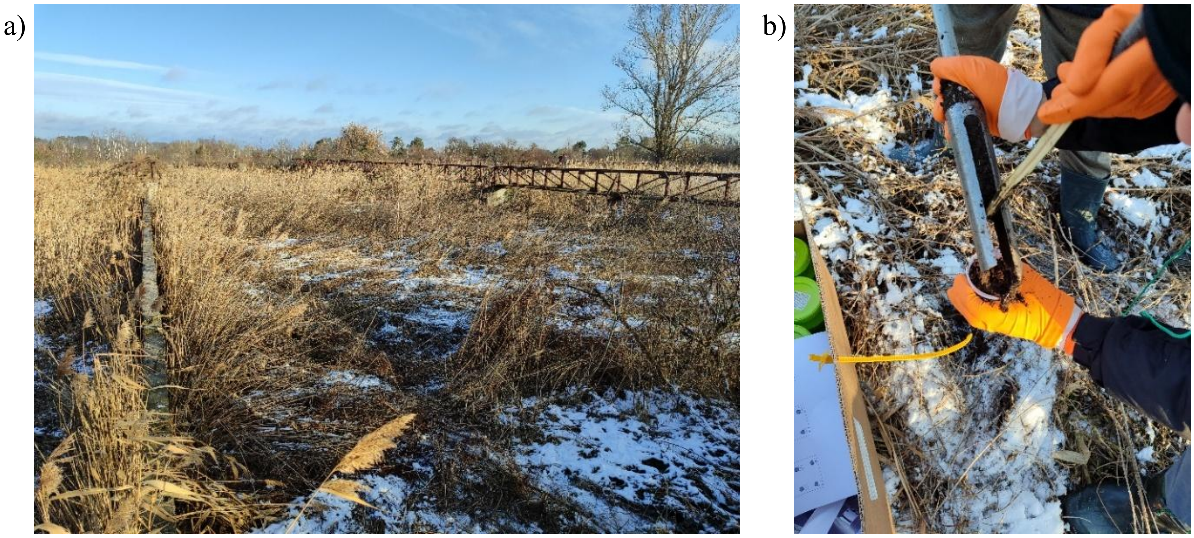

Fig. 1. **Sampling of sludge:** a – general view of the sludge storage area;
b – process of taking a sample using an improved grain sampling probe

Winter wheat straw has been selected as the biomass component, as it is a readily available agricultural raw material in all regions of the country. Its basic thermal characteristics (moisture content 12%, ash content 5%, net calorific value of dry matter 17.6 MJ·kg^{-1}) were adopted based on standard EN 14961-1:2010 and previous studies [11]. Taking into account the results of studies [15; 16], the lower heating value of the dry mass of the sludge was taken as 11.2 MJ·kg^{-1}.

The numerical experiment was carried out using a central composite design, which incorporates a full factorial experiment. Three key parameters were selected as independent variables (factors), with their levels of variation approximating real-world conditions (Table 1).

Table 1

Experimental factors and levels of variation

Experimental factors	**Levels of variation**		
	lower (-1)	**main (0)**	**upper (+1)**
X_1 – initial moisture content of the sludge, W_{SL}, %	60	70	80
X_2 – mass fraction of straw in the mixture, R_{ST}, %	20	40	60
X_3 – heavy metal (cadmium) content, Cd_{SL}, mg·kg^{-1}	1.5	4.25	7.0

The assessment of efficiency (per 1 kg of finished mixture) was carried out using the following criteria – response functions: mixture moisture content (Y_1, W_{MIX}, %, with a process constraint of $W_{MIX} \leq 35\%$); heat of combustion (Y_2, LHV_{MIX}, maximisation); cadmium content in the mixture (Y_3, Cd_{MIX}, minimisation); nitrogen-to-carbon ratio of the mixture (Y_4, C:N).

The response function values were calculated analytically. The moisture content of the two-component mixture W_{MIX}, % was determined using the additivity rule of the material balance [10] according to formula (1):

$$W_{MIX} = W_{SL}\left(1 - \frac{R_{ST}}{100}\right) + W_{ST}\frac{R_{ST}}{100}, \tag{1}$$

The lower heating value of the composite matrix (LHV_{MIX}) was estimated using the methodology set out in standard EN ISO 18125:2017, which takes into account the heat input required for the latent heat of vaporisation of water during combustion (2):

$$LHV_{MIX}\left[LHV_{SL}^{db}\left(1 - \frac{R_{ST}}{100}\right) + LHV_{SL}^{db}\frac{R_{ST}}{100}\right]\left(1 - \frac{W_{MIX}}{100}\right) - 0.02442W_{MIX}, \tag{2}$$

where LHV^{db} – lower calorific value of the component on a dry weight basis, MJ·kg^{-1};
0.02442 – heat of vaporisation of water, MJ·kg^{-1} per 1% moisture.

The response functions for cadmium content were calculated using the law of conservation of mass for dry matter (3):

$$C_{MIX} = \frac{C_{SL} m_{SL}^{dm} + C_{ST} m_{ST}^{dm}}{m_{SL}^{dm} + m_{ST}^{dm}}, \tag{3}$$

where C – concentration of the studied component;
m^{dm} – dry matter content of the relevant component per 1 kg of the mixture.

The statistical analysis of the results of the numerical experiment was carried out using Wolfram Mathematica and Microsoft Excel.

Results and discussion

The results of laboratory analyses of spot samples of sewage sludge taken from operational sludge storage sites formed the basis for mathematical modelling and the creation of an inventory database. The summary physico-chemical parameters are given in Table 2. The heavy metal content and acidity of the averaged samples for each of the three sites are given in Table 3.

Table 2

Physico-chemical parameters of sewage sludge

Sample	Moisture content, %	Dry matter content, %	Organic matter (carbon, C), %	Total nitrogen (N), % to raw material	Total nitrogen (N), % to dry matter	Carbon-to-nitrogen ratio, C/N
1 (11631/15)	71.88	28.12	23.02	1.00	3.57	6.448
2 (11631/16)	74.68	25.32	24.54	0.81	3.20	7.669
3 (11631/17)	73.95	26.05	20.79	0.71	2.74	7.588
4 (11631/18)	74.60	25.40	22.92	0.75	2.96	7.743
5 (11631/19)	76.84	23.16	26.41	0.93	4.03	6.553
7 (11631/8)	79.61	20.39	20.10	0.66	3.24	6.204
8 (11631/9)	84.36	15.64	28.05	0.72	4.59	6.111
9 (11631/10)	65.15	34.85	15.70	0.82	2.35	6.681
10 (11631/11)	80.25	19.75	28.32	0.77	3.91	7.243
11 (11631/12)	84.65	15.35	28.34	0.70	4.58	6.188
12 (11631/13)	79.25	20.75	26.61	0.77	3.69	7.211
13 (11631/2)	71.40	28.60	23.31	0.94	3.28	7.107
14 (11631/3)	78.60	21.40	27.01	0.94	4.39	6.153
15 (11631/1)	55.00	45.00	19.76	1.09	3.87	5.106
16 (11631/4)	71.55	28.45	22.43	0.92	3.22	6.966
17 (11631/5)	80.84	19.16	25.33	0.80	4.16	6.089
18 (11631/6)	79.38	20.62	26.03	0.82	3.99	6.524

Table 3

Heavy metal (cadmium) content and pH of the averaged sludge samples

Parameter	Sample		
	38 (11631/22)	39 (11631/14)	40 (11631/7)
Acidity, pH units	7.03	7.47	7.39
Mass fraction of cadmium (Cd) in dry matter, mg·kg^{-1}	6.93	1.65	3.97

Analysis of the data obtained confirms significant variability in the moisture content of the sludge, which is likely to depend on the depth of sampling and the intensity of natural drying. An average moisture content of 75.41% precludes direct pelletising or effective mono-incineration of the sludge. At the same time, the high organic matter content indicates significant energy potential. An important finding is the concentration of cadmium (max. 6.93 mg·kg^{-1}), which is several times lower than the strict limits of EU Directive 86/278/EEC (20-40 mg·kg^{-1}). This classifies the sludge from the studied treatment plants as an environmentally safe raw material: the ash residue after incineration will not require special disposal conditions, and in the case of bioconversion, the resulting fertiliser will be safe for agroecosystems. The nitrogen-to-carbon ratio of the sludge ranges from $C:N$ = 5.11 to 7.74 and does not show a functional dependence on moisture content. For further analysis, we take the average value of $C{:}N = 6.68$.

For comprehensive optimisation of the component composition, a Central Composite Design was implemented, comprising a full factorial experiment supplemented with star and zero points (a total of 15 experiments). The influence of the initial moisture content of the sludge (X_1), the mass fraction of straw in the mixture (X_2) and the initial cadmium content (X_3) on the response functions, calculated for a functional unit of 1 kg of the finished composite, was evaluated. The design matrix and the results of the calculations are presented in Table 4.

Table 4

Planning matrix and results of the numerical factorial experiment

No	Experimental design			Values of factors			Values of parameters			
	X_1	X_2	X_3	W_{SL}, %	R_{ST}, %	Cd_{SL}, mg·kg^{-1}	Y_1, W_{MIX}, %	Y_2, LHV_{MIX}	Y_3, Cd_{MIX}	Y_4, $C{:}N$
1	1	1	1	80	60	7.00	39.2	9.23	3.68	50.77
2	1	1	-1	80	60	1.50	39.2	9.23	0.79	50.77
3	1	-1	1	80	20	7.00	66.4	3.27	6.09	14.03
4	1	-1	-1	80	20	1.50	66.4	3.27	1.30	14.03
5	-1	1	1	60	60	7.00	31.2	10.32	4.83	28.72
6	-1	1	-1	60	60	1.50	31.2	10.32	1.03	28.72
7	-1	-1	1	60	20	7.00	50.4	5.45	6.51	10.35
8	-1	-1	-1	60	20	1.50	50.4	5.45	1.40	10.35
9	1	0	0	80	40	4.25	52.8	6.25	3.04	26.27
10	-1	0	0	60	40	4.25	40.8	7.89	3.54	16.48
11	0	1	0	70	60	4.25	35.2	9.78	2.66	36.07
12	0	-1	0	70	20	4.25	58.4	4.36	3.86	11.58
13	0	0	1	70	40	7.00	46.8	7.07	5.53	19.74
14	0	0	-1	70	40	1.50	46.8	7.07	1.18	19.74
15	0	0	0	70	40	4.25	46.8	7.07	3.36	19.74

Analysis of the process parameter (mixture moisture content, Y_1) indicates that the mass fraction of the added straw has a decisive influence on this parameter. For sludge with typical moisture content after mechanical dewatering (70%, experiments 11-15), the addition of straw (moisture content 12%) in a proportion of 60% by mass ($X_2 = 60\%$) allows the total moisture content of 1 kg of mixture to be reduced to the target value of 35.2%. This confirms the effectiveness of the "passive drying" method, which provides the necessary rheological properties for the operation of die-press pelletizers without the use of external thermal energy. If the sludge is pre-dried to 60% (experiments 5 and 6), the target moisture content of 31.2% is achieved even more easily. A moisture content of 30% allows for the use of pelletising technologies without pre-drying the raw materials. Thus [19], moisture loss in raw materials when using screw pelletisers can reach 30-50% of their initial content, whereas, for example, when producing compound feed on ring-die pellet mills with a feed mixture moisture content of 28-34%, the moisture content of the pellets at the die outlet is 20-25%, and after cooling in the cooling tower – 17-20%.

The energy criterion (Y_2) demonstrates that the sole incineration of raw sludge is inefficient due to the high costs associated with the latent heat of water vapourisation (for example, LHV_{MIX} drops to 2.5-3.2 MJ·kg^{-1} when the straw content is minimal, and the sludge moisture content is high). At the optimal ratio for pelletisation (sludge 40%, straw 60%; experiment No. 11), the calculated lower heating value of the working mass (LHV_{MIX}) reaches 9.78 MJ·kg^{-1}, and at a sludge moisture content of 60% – rises to 10.32 MJ·kg^{-1}. It is worth noting that these figures (9.7-10.3 MJ·kg^{-1}) reflect the energy value of the raw material mixture in its wet state prior to pressing. During the pretreatment of the raw material and pressing in the pelletiser die under the influence of temperature and pressure, the moisture content will decrease further, which is expected to increase the final calorific value of the finished solid pellets to 13-15 MJ·kg^{-1} [9; 17; 18]. These data require experimental verification; however, they correlate with data we obtained previously [12] regarding the calorific value of composite pellets based on sewage sludge and fallen maple leaves (50-70% by weight), which was in the range of 17.3-18.0 MJ·kg^{-1}. The results obtained also correlate closely with data from known studies [10], which recorded 11-16 MJ·kg^{-1} for finished briquettes made from similar components.

The change in heavy metal content (Y_3) clearly illustrates the "dilution effect". When clean biomass is added, the concentration of toxicants per unit of dry weight of the composite drops sharply. Even when using the most contaminated sludge (Cd_{SL} = 7.00 mg·kg^{-1}), adding up to 60% straw reduces the final cadmium concentration in the mixture to 3.68-4.83 mg·kg^{-1} (experiments 1 and 5). An additional advantage of this approach is a proportional reduction in the total ash content of the fuel, which significantly reduces the risk of slagging in heat-generating equipment.

Analysis of the results of the numerical experiment using regression and dispersion analysis methods enabled us to obtain adequate models of the influence of composite mixture parameters on the specified criteria (Table 1, 2). According to the results of the dispersion analysis, all obtained regression models are statistically adequate (confidence probability P = 0.95) and highly accurate – the models' coefficients of multiple determination $D \geq 0.99$. Tests using Fisher's F-test and Student's t-test confirmed the overall significance of the equations and their coefficients, respectively. For mixture moisture content (Y_1, W_{MIX}, %), an exponential model was obtained ($D = 0.993$, F = 890.2), which describes the multidirectional exponential influence of the analysed factors. Heat of combustion (Y_2, LHV_{MIX}) is reliably described by an incomplete polynomial model ($D = 1.000$, F = $6.7 \cdot 10^6$) with a predominance of linear effects and significant pairwise interactions between factors. The polynomial model for cadmium content in the mixture (Y_3, Cd_{MIX}) ($D = 0.998$, F = 699.7) is characterised by a significant ($P \geq 0.94$) antagonistic effect of pairwise interactions. For the nitrogen-to-carbon ratio of the mixture (Y_4, C:N), a full quadratic response surface was obtained ($D = 0.997$, $F = 573.2$), indicating the presence of a local extremum (minimum) due to highly significant quadratic effects of both factors.

Thus, the mathematical equations (4)-(7) obtained provide a reliable approximation of the experimental data and are suitable for predicting performance indicators and solving multi-criteria optimisation problems for the studied process.

$$W_{MIX} = 31.1012 \cdot 1.01274^{W_{SL}} \cdot 0.987482^{R_{ST}}. \tag{4}$$

$$LHV_{MIX} = 11.187 - 0.1363 W_{SL} + 0.040425 R_{ST} + 0.0013625 W_{SL} R_{ST} - 5 \cdot 10^{-6} R_{ST}^2. \tag{5}$$

$$\begin{aligned} Cd_{MIX} = {} & -1.8712 + 0.0214114 W_{SL} + 0.0612182 R_{ST} + 1.44464 Cd_{SL} - \\ & - 0.00054375 W_{SL} R_{ST} - 0.0055909 1 W_{SL} Cd_{SL} - 0.0002875 R_{ST}^2 - 0.00729545 R_{ST} Cd_{SL}. \end{aligned} \tag{6}$$

$$C:N = 125.927 - 3.001 W_{SL} - 1.80882 R_{ST} + 0.01925 W_{SL}^2 + 0.0229625 W_{SL} R_{ST} + 0.0109375 R_{ST}^2. \tag{7}$$

The results of the numerical experiment are consistent with previously reported trends in the co-pelletisation of sewage sludge with lignocellulosic biomass. In particular, moisture content and composition ratio are key parameters affecting pellet quality and energy consumption, and the use of straw as an additive is known to reduce moisture and improve fuel properties [20; 21].

The predicted increase in calorific value (10-13 MJ·kg^{-1}) agrees with literature data indicating that co-pelletisation with biomass enhances the energy characteristics and stability of sludge-based fuels [21; 22]. In addition, lignocellulosic components (e.g., lignin) and organic matter in sludge can act as natural binders, supporting pellet formation without additives [20]. At the same time, previous studies highlight that pelletisation energy demand depends strongly on process conditions, and that environmental aspects, particularly ash composition and potential heavy metal content, require further investigation [20-22]. Overall, the agreement with literature supports the validity of the proposed modelling approach; however, experimental validation remains necessary to confirm the predicted properties and environmental performance.

Based on the obtained results, a functional unit of 1 kg of composite pellets (wet basis) is proposed for further life cycle-oriented analysis. According to the developed mathematical model, a representative composition for subsequent assessment is a mixture of 0.4 kg of sewage sludge and 0.6 kg of wheat straw. This ratio provides a compromise between moisture reduction and calorific value improvement. In particular, the calculated lower heating value of the mixture reaches approximately 10-13 MJ·kg^{-1}, while the C:N ratio increases from an initial value of about 6.7 (raw sludge, Table 2) to approximately

25-30, which is favourable for biological processing (28.72 in experiments 5 and 6 of Table 4).

Within this framework, a simplified life cycle inventory approach was considered. The system boundaries include sludge handling, mixing, pelletisation and end-use via thermochemical conversion, while thermal drying is not required due to the structuring role of straw. The main input flows are sewage sludge, straw and electricity for pelletisation, whereas the outputs include composite pellets, ash and energy release during combustion.

A simplified energy balance indicates that the specific energy demand for pelletisation (typically 0.05-0.15 kWh·kg^{-1}) represents only a small fraction of the energy content of the produced fuel. Therefore, the proposed approach may provide a potentially positive energy balance under the assumed conditions. At the same time, environmental performance, including emissions and ash safety, requires further investigation and cannot be conclusively assessed within the scope of the present study.

Conclusions

1. The direct use of sewage sludge as a fuel is limited by its high moisture content (on average 75.41%), which negatively affects its energy performance. The addition of approximately 60% dry wheat straw allows the moisture content of the mixture to be reduced to about 31-35%, which may correspond to a technologically acceptable range without the need for additional thermal drying.
2. The lower heating value (LHV) of the raw material mixture with a representative composition (40% sludge and 60% straw) is estimated at 9.78-10.32 MJ·kg^{-1}. Based on the developed model, pelletisation with moisture reduction may increase the effective LHV to approximately 13-15 MJ·kg^{-1}, which is comparable to typical values reported for biomass-based solid fuels.
3. The sewage sludge from the studied wastewater treatment plants has a low level of heavy metal contamination (maximum 6.93 mg·kg^{-1} of cadmium). The addition of straw leads to a dilution effect, reducing the cadmium concentration in the mixture to 3.68-4.83 mg·kg^{-1}.
4. A representative substrate composition (0.4 kg of sewage sludge and 0.6 kg of wheat straw per 1 kg of mixture) is proposed as a functional unit for further analysis. This composition improves moisture content and C:N ratio (to approximately 25-30:1) and can be used as a basis for simplified life cycle inventory and subsequent comparative assessment of sludge utilisation pathways.

Author contributions

Conceptualization, V.B.; methodology, V.B., V.K and V.R.; validation, V.B., V.R., O.A.; investigation, V.B., V.K., V.R., O.A., S.K.; data curation, V.B., V.K., O.A., S.K.; writing – original draft preparation, V.B., V.K.; writing – review and editing, V.B., V.K., V.R., O.A., S.K.; visualization, V.R., O.A., S.K.; project administration, V.B.; funding acquisition, V.B. All authors have read and agreed to the published version of the manuscript.

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