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Review

Granulates Based on Bio and Industrial Waste and Biochar in a Sustainable Economy

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Abstract: This review presents the latest research works detailing granulation processes and granulates, including and based on waste (bio and industrial) as a biofuel/energy source and the possible usage of granulates from and/or based on biochar. The innovative aspect is that the article focuses on the broadest possible environmental aspect understood in minimizing the burden related to the amount and composition of waste generated by various industries. The aim of the study is to demonstrate the processes as an effective method of waste management and also as energy sources. Based on various sources, a brief summary of why granulation is an important area of both scientific research and industrial applications is provided. The review also presents a summary of basic concepts and definitions in the topic of granulation—types of processes, apparatuses used, and examples of research results in the literature. The main part of the review is the analysis of the literature providing numerous examples on the usage of granules based on bio and industrial waste and various biochar granulates. The conclusions present the aspect of economical sustainability of granulation processes and the use of granulates as effective solutions for energy sources (fuel, biofuel), waste management, and applications in agriculture (soil additives, fertilizers).

Keywords: granulation; granules; biomass waste; industrial waste; biochar; biofuels; soil additives; waste management; sustainability

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1. Introduction

The Polish energy sector is facing considerable challenges. The key difficulties confronting this industry include excessive energy consumption, insufficient infrastructure development, fuel and energy production and transportation, extraction of foreign energy natural gas extraction, and rising CO_2 pricing [1,2]. Because of the generally recognized demand to reduce carbon dioxide emissions, biomass, especially waste biomass from the agriculture and food sector, has been employed more frequently as a source of heat and power generation in the past few years [3,4]. Wood pellets are one of the most extensively utilized biofuels because they are inexpensive, homogeneous, and simple. Given the increased interest in this type of resource, which includes industrial and biological waste, various approaches for improving biofuel quality with granulation procedures are being researched.

Agglomeration, granulation, and pelletization are the currently available industrial-size expansion methods. Granulation and agglomeration are synonyms for the particle manufacturing or size expansion process, which encompasses a wide variety of procedures used to create agglomerates of varying dimensions in which the original particles can still be recognized, with the latter process producing particles that are larger and more diverse in size while also requiring the mixing of materials. The granulation method is one of the viable densification process alternatives. Recent research has investigated the influence of material qualities on granulation, both with and without binding additives. Binders have been used to decrease biomass elasticity and maintain the highest bulk density. Various granulating agents such as lignin, bio-oil, and beer waste, but also cellulose-based compounds (such as methyl or ethyl cellulose), gum acacia, starch, and modified starch, were used as binder materials at different rates [5].

Compared to pressure agglomeration, non-pressure granulation can reduce energy usage and allow for simpler scaling up of the process, also improving the flowability and bulk density of biomass. Granulation is commonly applied in mineral processing as well as in the manufacturing of detergents, cosmetics, food, energy sources, and fertilizers, and also of pharmaceuticals and excipients [6].

The aim of the study is to indicate possible methods of material processing, including waste, with granulation methods (dry and wet) used, inter alia, as a source of energy. Granulation is an effective and environmentally friendly process, i.e., waste-free. Additionally, the produced granules in their composition may contain waste residues generated from various industries. This article is part of the work conducted by the Łukasiewicz Research Network–Lodz Institute of Technology on the migration of components from granules produced by non-pressure methods obtained from environmentally harmful waste (including waste generated by leather and tanning, agri-food, mining, chemical, and other industries).

This paper's innovation focuses on the broadest possible environmental aspect understood in minimizing the burden related to the amount and composition of waste generated by various industries. Moreover, appreciating the achievements of the authors (predecessors) of research on this subject, it should be stated that the discussed analyses go beyond the source articles' subject matter. The paper also focuses on the rarely studied aspect of using granulation processes as an effective technology in the field of waste management (especially of industrial waste) and the processing of various types of biomass waste.

2. Materials and Methods

Granulation Types

Dry and wet granulation techniques are the two main branches of the granulation process.

2.1. Wet Granulation

The procedure adheres powder particles together with the usage of various liquid-binding additives which form granules due to the creation of connections between grains. The liquid is fed and dispersed to the grains by the granulator's mixing. Capillary pressure and surface tension forces between liquid particles (liquid bridges) are the leading causes of granule formation and strength [7]. It is a promising method for producing cosmetics, pharma and biologic tablets, nutraceuticals, agriculture, and food ingredients [8].

The wet granulation process has three fundamental stages, as described below [9].

Wetting and nucleation: Granulation begins with particle attachment caused by additive bridges, thus creating agglomerates at capillary states which can serve as small aggregates (nuclei) for subsequent granule expansion.

Consolidation and growth: There are two ways of consolidation. Single particles start to link with the core through pendulum bridges, or nuclei particles start to merge and nuclei begin to grow.

Breakage and attrition: The creation of tiny particles distinguishes it as a result of granulator impacts and product processing or granule rupture. All wet granulation procedures have these three mechanisms. The first part of the process is spraying, when particles are sprayed with water or a binder solution. The second phase is agglomeration, when particles collide and begin to stick together. The last phase is finished granulate, when particles remain bound after agglomeration.

Various types of granule-producing apparatuses are mostly used in wet granulation, such as:

Fluidized-bed granulators: Proper gas-distributor design is essential to sustain solids mixing and heat and mass transport. In this type of machinery, an aqueous or solvent-based liquid binder is sprayed by an atomizing, two-fluid nozzle which could be placed in various positions around the bed. Parameters such as spray distribution, atomizer design, and humidity management are all critical to efficient functioning.

Tumbling granulators: A continuous, inclined disc and drum granulators with 1–100 ton/h efficiency and residence durations of 1–5 min are common designs. Disc granulators are comprised of a revolving pan with a rim that is slanted at horizontal angles of 50–60 degrees. Drums are cylindrical, with a horizontal angle of 3–10 degrees and can have annular retaining rings.

Mixer granulators: Mechanical agitation causes granulation in mixer granulators. A broad range of mixing-tool designs are offered. Manufacturers' shear rates and powder-flow patterns vary greatly due to equipment, impeller, and chopper geometry differences. Agglomerate size and density are determined by control of the liquid phase, wet mass rheology, and the intensity and duration of mixing.

2.2. Dry Granulation

The process is accomplished using roller compaction or slugging methods. Dry granulation uses mechanical pressure to encourage the agglomeration of dry powder particles, whereas wet granulation combines raw material powder with granulation solution to generate wet matter and enhance agglomeration. This process mainly uses roller compaction granulation instead of wet granulation which uses fluidized-bed, high-shear, or twin-screw granulation [10].

Using a spinning disc/cup is more efficient in this procedure compared to other techniques. However, the process has not been commercialized due to some design challenges to the granulator and disc design that would prevent reheating of the collected granules and, as a result, their sticking together. Shanmugam [11] produced a schematic representation of each granulation method and described current advances in granulation technology.

3. Results

Table 1 presents a list of exemplary granulation processes, including, e.g., biomass. The obtained granules were characterized by grain fineness of 0.24–18 mm and the potential to be used as solid biofuel. This method of rainfall processing allowed, among others:

✓ Increasing the reactivity of fuel;
✓ Improvement in flame temperature and boiler efficiency;
✓ Higher energy potential;
✓ Enhanced sinter output and other.

Table 1. Examples of granulation research in the literature (Web of Science Core).

Biomass/Material and Process	Granulator Type	Size	Result	Reference
Iron ore, coke Iron sintering	Wet granulation, long inclined rotating drum	0.25–1.0 mm	The preferred positioning of coke particles on the exterior of granules, as well as modifying their size distribution, improves gas access to the granulated material, increasing the reactivity of fuel.	[12,13]
Agricultural bio-waste (cow slurry, grass silage digestate, chicken litter) Combustion	Wet granulation—PVC drum at room temperature	<3.15 mm	The study compared the performance of granulated agricultural bio-waste as a solid biofuel throughout the combustion process to two typical wood fuels. Between granulated different bio-waste and non-granulated wood fuels, combustion studies indicated substantial improvements in flame temperature and boiler efficiency.	[14]

Table 1. *Cont.*

Biomass/Material and Process	Granulator Type	Size	Result	Reference
Liquid fraction of anaerobic digestate with limestone powder Pelletization	High-shear wet granulation—Kenwood-KM070 (planetary mixing)	2–4 mm	The liquid-to-solid ratio increased during granulation, resulting in higher granule strength and product yield. Though chemically synthesized fertilizer granules have slightly higher strength (approximately 5 MPa to 7 MPa) than resulting granules, this can be addressed by adding polymeric binder or dusting the particles at the end of the process.	[15]
Compost—mixture of food scraps, dry leaves, wood chips, stones, straw, etc. Combustion	Wet drum granulator	3.35–14 mm	Drum granulator was used to effectively densify compost bio-waste into granules, with sodium silicate as a low-cost binder to allow easy storage and transportation. Presented research has shown that silicate-based binders may be utilized to manufacture high-quality bio-waste granules which have the potential as a solid biofuel.	[16]
Fibrous hemps (Felina 32, USO 31, Finola) Combustion	Horizontal granulator, Polexim pelleting machine	2–6 mm	The study proves that pellets made from fibrous plants have the potential to be used as fuel while being an alternative for wood, oil, or gas.	[17]
Softwood and wheat straw	KAHL 14–175 pelleting press	6.0 mm	The HHV and LHV results for all granulated samples show that granules made from both types of biomasses had higher energy potential than the original samples.	[18]
Quasi-fuel particles, biomass char Iron sintering	Wet drum granulator	5 mm	Granulation of biomass char with an additive of 2% magnetite concentrate resulted in better parameters, such as tumbler strength (from 44.9% to 55.0%), sinter output (from 41.9% to 49.3%), and productivity (from 28.54 $t/m^2/d$ to 31.10 $t/m^2/d$).	[19]
Reed biomass Pelletization	-	-	Using potato starch as a binder in manufacturing pellets for fuel gives the best results, improving their quality, because of spreading uniformly throughout the granulated material and enhancing its capacity to agglomerate while also minimizing friction losses.	[20]
Onion husk waste Pelletization and combustion	Flat matrix- thickening roller P-300 pressure granulator	1.0–4.0 mm	In the study, the best quality granulate was obtained from a combination comprising 10% potato pulp, compacted in a 170 rpm matrix, resulting in an approximate density of 650 kg/m^3 and kinetic strength of 99.50%.	[21]
Sewage sludge + (coal slime, sawdust, and bone meal) Pelletization	Drum granulator	1.5–3.5 mm	When sewage sludge was turned into pelletized biofuel, typical issues relating to the co-combustion of dry sludge with coal were avoided. The highest CO (230 mg/s) and CO_2 (5000 mg/s) emissions were found on bone meal + sewage sludge combustion.	[22]
Carbonation lime mud	Wet disc granulation	4, 5, 6, 3, 8, and 10 mm	Sugar waste in the form of carbonation lime mud can be granulated and used as a soil de-acidifying fertilizer, because of its chemical content and the addition of additives. Instead of causing environmental problems with dusty waste, it can be commercialized and sold, bringing profit to the companies.	[23]

Table 1. *Cont.*

Biomass/Material and Process	Granulator Type	Size	Result	Reference
Different wood pellets + waste sunflower cooking oil treatment Upgradation of pellets	Ready commercial pellets were used	6–8 mm	Waste cooking oil has been added to the wood pellets at rates from 2% to 12%, in relation to the weight of the pellets. As a result, pellets with increased calorific content were obtained, without reducing their durability. On average, the maximum dose of the modifier (12%) resulted in a 12–16% increase in calorific value. Moreover, a reduction of ash concentration was obtained in all samples, with an average drop of 16–38%.	[24]
Post-harvest sage waste + rye bran Combustion	P-300 granulator, flat matrix-compacting roller		The researchers added (10% to 20%) rye bran to the granulation process of post-harvest sage waste to increase the quality of granules as fuel. The inclusion of rye bran reduces the granulator's power need (from 3.75 kW to 3.19 kW) and its physical and bulk density, increasing its kinetic durability. The resulting granules have a heating value (at 10% moisture content) ranging from 18.17 MJ/kg to 19.39 MJ/kg. Adding 20% rye bran reduces the latter by 2.07%, while at the same time, it increases the first one by 2.67%.	[25]
Waste tannery shavings + mineral additives (wet gypsum, dolomite)	Wet pulp granulation—vibrating disc granulator	less than 1.0 mm—more than 14.0 mm	More persistent, mechanically stable granules were obtained during the granulation process using various additives (water glass, dolomite, gypsum). Granules with grain size > 14 mm had the highest percentage.	[26]
Coconut shells Combustion	Pressured 37 MPa and 37 MPa granulation	8.0–12.0 mm	Obtained granules have a humidity of 9.4% combined with low ash content (0.66%). They also have a high calorific value (17.31 MJ/g) and a high volatile content (77.7%). The research discovered that using 47 MPa pressure agglomeration on ground granulates (8 mm diameter sieves) resulted in parameters of the highest mechanical durability with specific density, which were equivalent to market-available biofuels.	[27]
Sawdust residues + rye bran Pelletization	P-300 granulator, flat matrix-compacting roller		According to studies, adding rye bran to sawdust increases the susceptibility of the test mixture to densification, decreases energy consumption, and decreases the moisture with increasing rye bran content. With the addition of rye bran, the kinetic strength of granules increased from 94% to 98%.	[28]
Side, elephant grass, and reed canary grass Combustion	Horizontal mixing granulator	6.0 mm	Calorific values were found to be 17.4, 17.8, and 17.4 MJ/kg, respectively. It was determined that using a dryer to dry biomass resulted in marginally higher granules price (0.13 EUR/kg) compared to non-dried biomass (0.12 EUR/kg).	[29]
Switchgrass + lime pretreatment	Pan granulator—DP-14 agglomiser	1.0–18.0 mm	Untreated granules had higher bulk density than lime-treated granules. Lime treatment enhanced single granule density somewhat but lowered granule hardness, resulting in considerable susceptibility to fracture. Moreover, lime treatment raised the ash content while lowering the heating value of the granulate, thus being less suitable for energy and heat sources.	[30]

More research can be found in the literature also focusing on the granulation of bio and agricultural waste such as sewage sludge, rice straw, pine wood powders, herbaceous plants, wood shavings, and industrial waste such as spent malt, lightweight aggregates, coke, and biochar, to produce more economically and environmentally efficient biofuels, improve energy production from other solid fuels, or as a method of effective waste management. Biofuel by-products such as ash can be used as an additive in the agriculture sector for fertilizer production. In the experiment conducted by scientists from the Magnus University Agriculture Academy, a 25% increase in the concentration of biofuel ash in poultry manure had a positive effect on the increase of granules strength to 55% [31]. Nazarov et al. conducted studies of various biofuel granulates based on biomass waste, i.e., wood shavings, spent malt, peat, and sunflower husks, among others. Performed experiments showed that, depending on granulate composition, obtained agglomerates had a calorific value from 15,300 J/g to 22,315 J/g, with ash content from 3.5 to even 10%, which are comparable to other fuels (coke, coal) [32]. Other researchers, such as Jasinskas et al., used pellets from coarse herbaceous plants. Three types of energy plants were used—miscanthus, sida, and cup plant. Studies showed that the most critical parameters, i.e., carbon content, calorific value, and harmful gas emissions, have met the requirement for use as a high-quality biofuel [33]. Another study, conducted by Yilmaz et al., showed that pellets made from agriculture biomass waste and sewage sludge have sufficiently high energetic parameters to be used in co-combustion with other solid fuels such as coal. Compared to traditional fuels, such granules are easier to transport and store [34]. Yandapalli et al. showed that the agglomeration process can produce good-quality pellets from pine wood powders as a method of efficient waste management but, more importantly, as a potential cost-competitive biofuel [35]. Another study by Wzorek showed that various, dried mixtures of sewage sludge can be used as biofuel, even with low calorific value. It should be noted, however, that it is necessary to use other, properly selected components for a satisfying economical effect [36]. Tan et al. used plant-based biomass material in the form of rice straw in exploring new compression molding methods to further improve its potential, such as briquetted biofuel [37]. Zhou et al. performed an experiment in which they used a pellet mixture of coke and biomass char to investigate its combustion properties. Research has shown that such biofuel has high efficiency, and by increasing the coarseness of the pellets, with an additive of $Cu_{0.1}Ce_{0.9}O_2$ (3 wt.%), it is possible to improve the effectiveness of the combustion process of ~98%, while also increasing the replacement rate of biomass for coke [38]. Yliniemi et al. experimented with using ash from the fluidized-bed combustion process to produce lightweight aggregates as an effective method of waste utilization. Fly ash granulates were produced using a high-shear granulator with various binders. Obtained granules have physical properties which are no worse than lightweight aggregates produced with other methods. Moreover, it is possible to further increase their parameters by using various binders, such as blast furnace slag or metakaolin [39]. Ma et al. used a granulation process combined with drying to substantially reduce water content in sewage sludge and to co-incinerate it with coal, thus reducing fossil fuel consumption and CO_2 emission [40]. Skwarek et al. showed that the granulating process can be used as an effective seed-coating method. Various coating materials (two types of waste collagen, PHMB and waste dolomite) have been used on pea seeds to form granules, with positive effects on plant growth observed [41]. In another study, Ławińska proved that the granulation process can be used in seed coating with collagen and/or keratine (by-products from the leather industry) preparations as biofertilizers and foliar fertilizers [42]. Obidziński et al. conducted research that confirmed the possibility of using granules from food industry waste as components for producing pellets as groundbait for fish [43].

4. Discussion

A discussion on waste agglomerates as an energy source is presented below. The first part presents the concepts and examples of bio-waste and industrial waste (molten slag). Non-pressure and pressure granulation processes are widely used as waste management methods in various industries, including mineral processing [44,45], agriculture and forestry [46], energy [47], leather production [48], fertilizer production [26,49], and the sugar industry [23]. In recent years, scientists have increasingly researched the possibility of using waste as an energy source using granulation processes [50]. The following part of the article reviews the research results in waste agglomerates which can be used as fuel/biofuel. Agglomerates have been divided depending on the type of waste or granulation methods used.

4.1. Bio-Waste

Bio-waste understood as waste of plant and organic origin, such as:

- Forestry waste (needles, brushwood, bark, cuttings, etc.);
- Waste from the wood industry (sawdust, dust, shavings, etc.);
- Residues of agricultural activity (leaves of arable crops, silage), cultivation of energy crops (rape, flax, hemp and other oil plants, willow, miscanthus);
- Organic waste (liquid manure, manure, molasses, fruit pomace, and slaughterhouse waste).

They can be used, after appropriate processing, as biofuels or their components. For their production, pressure granulation is most often used, with the end product pellets or granules, which have been described in numerous scientific publications. Hejft and Obidziński tested the process of briquetting and pelletizing biomass waste in the form of buckwheat hulls with added potato pulp content (15, 20, 25%). The results have shown that with an increase of potato pulp content in briquettes and pellets, average kinetic durability decreases (from 94.8% to 80.9% for briquettes and from 98.2% to 91.4% for pellets) while also reducing average energy consumption by 46% [51]. In another work, Skonecki and Portręć studied the effect of humidity on pellets made from plant biomass (miscanthus, prairie spartin, and Virginia mallow). The results proved that the increase in raw material moisture positively affects its susceptibility to densification and the agglomeration process. It also decreases the average energy demand of the process (within the humidity range of 10–22%) by 14.7%, which positively affects the economic costs of agglomeration. On the other hand, increasing moisture content decreases the mechanical resistance of the agglomerates by 25% on average, which means that they are more susceptible to falling apart during storage [52]. Skonecki et al. also studied the relationship between moisture content and the susceptibility of materials of plant origin (wheat meal) to the pressure granulation process. The result indicated that a higher moisture level can increase the susceptibility to compaction of the tested material. Higher compaction means that it is possible to produce solid biofuel in a more sustainable process [53]. Shaw experimented on two biomass materials (poplar wood and wheat straw), studying the effects on their densification process and the possibility of producing high-quality pellets. The research showed that the increased pressure in the pelletizer resulted in an initial increased density of the pellets. The same effect was achieved by the increase in die temperature, while it also increased the tensile strength of the pellets. Experimenting with feedstock particle size has shown that decreasing it increases the density and tensile strength of the pellets. Moreover, as cited in the above papers, a decrease in moisture content resulted in higher granulates density. The study demonstrated that by pretreating biomass, it is possible to receive the end product (for example, granulated solid biofuel) which has higher quality and better parameters than untreated biomass material [54]. Niedziółka et al. researched the impact of different biomass pellets composition on the power consumption of an electric engine powering a pelletizer. The research demonstrated that biomass mixture does have an effect on energy usage, and also the amount of ash from the combustion of tested granulates [55]. Kaliyan and Morey tested various parameters of the pressure granulation process for biomass products and non-productive external factors. The results of their research showed that not only mixtures or extractives of biomass can have an effect on the increase of the density and durability of the final products (pellets, briquettes) but also storage conditions, i.e., temperature or humidity [56]. Szpryngiel et al. studied bulk density and energy usage of pellets production (from cereal straw and meadow hay) using pressure granulation. The results showed that it is possible to reduce energy usage of the granulation process by increasing the rotational speed of the densifying die in the pelletizer by 12.5% for straw and 15% for hay. On the other hand, the pelletizer used in the research required the use of biomass material with a moisture content above 25%, which means that obtained pellets need to be dried prior to their storage [57]. Niedziółka et al. researched the energetic and mechanical properties of pellets made from plant-based biomass waste—straw of various origin (wheat, rape, maize) was used. The researchers tested the energetic value of the pellets, which was in the range of 15.33–16.22 MJ kg^{-1}. The highest tested bulk density was for a rape–wheat mixture (523.6 kg m^{-3}) and for maize straw (566.9 kg m^{-3}). As for mechanical endurance, the highest values were recorded for maize and wheat straw (97.6%) and maize straw and wheat straw (97.6%). Those agglomerates have met European standards for solid biofuel pellets [58]. Miranda et al. reviewed the characteristics of pellets made from woody biomass of various origin—forest waste, wood industry waste, and woody agricultural waste, and compared the results with limit values by the EN ISO 17225 standard. The study showed that pellet granulated from pine sawdust from the sawmill industry was close to meeting the standard requirements, although nitrogen and ash values were exceeded. Other pellet granulates, such as pyrenean oak, pyrenean sylvestris, and powder from the cork industry, met most of the standard requirements, particularly concerning moisture, bulk density, and mechanical durability. It should be noted that almost all analyzed pellet granulates had heating value exceeding the standard's upper limit. The study showed that granulates from woody biomass could be used as solid biofuels in commercial and residential (pine sawdust) or industrial applications (remaining pellets) [59].

One example is the analysis of the use of oak waste as a source for the production of granular biofuels, the production of thermal energy, or energy conversion, presented by Jasinskas et al. Pellets made of oak waste (bark, leaves, and their mixtures) have a high calorific value (17.7 MJ kg^{-1} DM) and also generate lower emission values of harmful gases (CO_2, NO_x) than, for example, straw pellets (4–5 times lower emission CO_2) or peat. Combustion parameters such as ash content, for example, are at a lower level than conventional biofuels, but the overall results of combustion and emission studies indicate that granulation and additional preliminary operations (grinding, drying) can be used as biofuels [60].

Another example of the use of waste biomass of plant origin is provided by Chen et al., who showed that in the wet granulation process with the addition of sodium silicate as a binder, it is possible to produce, on a laboratory scale, biofuel pellets based on compost with a size from 3.35 mm to 14 mm, with properties not worse than fuels from wood biomass [16].

Jasinskas et al. analyzed the waste from broad bean cultivation in terms of its possible use as an alternative to fossil and wood fuels. The waste from broad beans was processed into cylindrical pellets as part of the research. The calorific value of the pellets obtained in this way ranged from 16.9 MJ kg^{-1} DM to 17.1 MJ kg^{-1} DM, which is a result comparable to some pellets made from the wood industry and forestry waste. In addition, an analysis of harmful gas emissions was conducted, i.e., carbon monoxide CO, carbon dioxide CO_2, unburned C_xH_y hydrocarbons, and nitrogen oxides NO_x, and they did not exceed the permissible limits. For some samples obtained from cultivation using the technology of untreated soil, they were even two times lower than, for example, conventional plowing. Thus, the obtained results confirmed the possibility of using pellets from waste as a source of biofuel [61].

Chen et al. assessed the possibility of using agricultural bio-waste as a potential biofuel and compared the granules produced with the use of a drum granulator with wood fuels. Research has proved that this type of bio-waste can be an alternative to solid fuels burnt in multi-fuel boilers. The best results were obtained by burning granules based on cow slurry with grass silage digestate, as well as the mix of wood industry and forestry waste, chicken litter. and digestate. They had the highest combustion temperature and met the energy requirements for biomass fuels [14].

The literature on the subject also includes studies on the suitability of granules produced in a drum granulator based on municipal sewage sludge for co-combustion with coal in grate boilers. Three types of pellets with a grain size of 15 mm and 35 mm were tested:

- Mixed sewage sludge with coal sludge;
- Mixed sewage sludge with meat and bone meal;
- Mixed sewage sludge and sawdust.

Quick lime was an addition to each biofuel (from 1 wt.% to 6 wt.%). Laboratory tests have shown that biofuels prepared in this way have comparable combustion rates and emissions of CO_2, NO, and SO_2 as solid fuels based on coal. The problem was the increased emission of NO_x and the deposition of residues after the combustion process (ash, slag) in a laboratory installation simulating water boilers with stationary and mechanical grates. The authors note, however, that pelletized biofuel based on sewage sludge is a suitable method for the management of this type of waste, because the composition of the granules is similar to pea coal and allows them to be co-incinerated in grate furnaces. Wzorek evaluated the potential of biofuels based on municipal sewage sludge (MSS) in such applications. Three types of biofuel mixtures were studied. The first mixture consisted of sewage sludge and coal slime—PBS fuel. The second consisted of sewage sludge and meat and bone meal—PBM fuel. The third consisted of sewage sludge and sawdust—PBT. Each fuel has been in two granulates sizes—15 mm and 35 mm. The conducted test on laboratory installation which simulated combustion in a stoker-fired boiler showed that sewage sludge-based biofuels have the potential to be used in co-combustion with coal in grate furnaces [22]. In another study, Yilmaz et al. produced pellets from a mixture of sewage sludge and animal and agricultural (olives) waste using pressure granulation. Analysis of the obtained biofuels included both the physical parameters and energetic properties. Tests have shown that the obtained granulated biofuel pellets have adequate properties to enable storage and transport. The only disadvantage is high water absorption, so they should be stored indoors in dry conditions. Tests of energetic properties have shown that analyzed biofuel has the potential to be used in the process of co-combustion with coal or in industrial applications such as the clinker burning process [34]. Wzorek analyzed a granulated mixture of municipal sewage sludge with meat and bone meal using a drum granulator. The test showed that energetic parameters such as low heat value, moisture content, volatile matter, and ash content met the requirements of the cement industry for alternative fuels. The disadvantage of such a biofuel granulate was the high content of P_2O_5, which cannot exceed the required values. Another disadvantage was the high water absorption of the obtained granulates, which means that dry storage is needed. On the other hand, the properties of tested granulates show that there is the potential for using them in power- or heat-generating plants in co-combustion with coal [62]. Wzorek and Głowacki also developed a device for mixing materials, especially sewage sludge, for use in research concerning the granulation of biomass [63]. In another study of biomass usage in co-combustion, Pronobis studied the effect of biofuels made from dried sewage sludge, straw, and wood on boiler fouling. The results of the tests showed that by using granulated biofuels, there is the potential to reduce the usage of coal with satisfactory boiler efficiency loss. However, the challenge is to significantly lower biofuel prices and use soot blowing to eliminate stronger boiler fouling while using biomass-based biofuels [64]. Garcia-Maraver et al. also analyzed biomass biofuels, i.e., sewage sludge, for biomass ash deposition and fouling tendency. The result of their research showed that while it is one of the barriers for use as a solid fuel, there is a need for further research on creating and validating indices for different biomass-based fuels, combustion technologies, and operating conditions [65].

Arshanitsa et al. investigated the possibility of using non-hydrolyzed wheat straw residues in the ethanol production process as potential granular biofuels. The work showed that due to the higher carbon content, unhydrolyzed wheat straw (LHR) thus has a higher calorific content than wheat straw, although slightly lower than coniferous wood. In addition, the waste material, thanks to its thermoplasticity, is easier to granulate and has greater efficiency than other biomass fallout, i.e., softwood sawdust. The finished LHR granules also had lower requirements for additional thermal energy, and thus achieved a lower flash point than softwood granules. Due to the higher emission values of nitrogen oxides NOx and carbon monoxide CO, the recommended solution is to use granules in co-combustion processes, e.g., with propane, which allows a reduction of CO emissions and at the same time increases the total heat output by 10% [66].

Gageanu et al. showed that granulation processes can have a positive effect on the possibility of using waste plant biomass with a lignocellulosic structure (wood, straw, sawdust, tree bark, etc.) as energy sources. As part of their research, Salix viminalis willow sawdust granulates with three different grain sizes were prepared using a 100 kN pressure granulator. The granulation process made it possible to eliminate the greatest disadvantage of sawdust materials, i.e., low density, which thus leads to difficulties in their transport and storage, and at the same time increases economic costs. Studies have shown that the granulation process does not adversely affect parameters such as calorific value or ash content, which makes it possible to recommend this process for use in the production of biofuels from lignocellulosic materials [67].

Mudryk et al. proved that granulation processes can have a positive impact on the effects of burning biofuels based on plant biomass. A common problem in the combustion processes of this type of material is the phenomena of ash fusion (slagging) due to the content of sulfur that precipitates during combustion (as sulfur oxides), which creates a high risk of corrosion of the heating surfaces of biomass boilers. During the research, a 3% kaolinite addition was used in the granulation process, effectively preventing ash from melting in various temperatures (from 900 °C to 1100 °C). It is therefore possible to apply the addition mentioned above in various biofuels based on plant biomass without the risk of the corrosion of heating boilers [68].

Jasinskas et al. studied fiber plants in terms of the possibility of using granulated, pelleted biofuel based on the plants for use in combustion and energy generation processes. The results of the research confirmed that the tested fiber plants, i.e., three types of hemp and one of nettle, have the potential to be used as biofuels. The obtained pellet was characterized by a relatively low humidity (from 8.87% to 9.98%), a relatively low ash content (3.58% for hemp and 6.6% for nettle), and a fairly high net calorific value—17.37 MJ kg^{-1} (hemp) and 16.93 MJ kg^{-1} (nettle). Tests were also performed for the emission of harmful gases (CO_2, CO, NOx, unburned CxHy hydrocarbons)—all values were within acceptable limits. The research also showed that thanks to additional preliminary operations, i.e., drying the material and reducing the humidity by 20%, it was possible to achieve the energy efficiency coefficient of 2.83 [17].

The presented examples showed that the granules based on biomass have great potential for biofuel use. In addition, granulation processes have a positive effect on the combustion effects of biomass-based biofuels. However, it should be noted that issues such as using municipal waste and biomass from municipal biological waste as biofuel or solid fuels components should be expanded in subsequent works. Moreover, some of the logistical and economic issues of bio-energy fuels will be covered in future papers. This article did not focus specifically on these issues, as the authors' aim was to present environmental issues as broadly as possible, i.e., to minimize the burden related to the amount and composition of waste generated by various industries.

4.2. Industrial Waste—Molten Slag

The metallurgy sector produces various types of by-products, with one being slags. The standard technology for the management of this waste was water quenching, but it is a process in which the potential for recovering waste energy is significantly limited, mainly because of its high operating temperatures, ranging from 1250 °C to 1650 °C. In addition, this process generates harmful gaseous waste such as SOx and H_2S that are released into the environment. Therefore, due to new environmental protection policies and constantly increasing energy costs, scientists began research on the development of an effective technology for waste heat recovery using the dry granulation process [69,70]. Different groups of researchers approached this issue in different ways. Luo et al. conducted research on hot slag particles as a heat transfer medium to produce hydrogen-rich gas by bringing biomass into a gaseous state using a moving-bed reactor [71]. Sun et al. developed an installation using hot slag particles and consisting of a rotary cup atomizer and gasifying system for CO_2/sludge gasification [72]. Liu et al. used an installation consisting of a rotating cup with a gravity-bed waste heat boiler. Molten slag flowed through the rotating cup, then fell into the boiler and circulated through the boiler pipes, being gravity driven. In the last stage, heat conversion from slag particles to a sensible heat of water took place [73]. Li et al. investigated the possibility of using solid slag for coal gasification, in which CO_2 was a gasifying agent in a fixed-bed reactor [74].

The common feature of all the above studies was the use of a rotating cup or disk, into which molten slag was poured and subjected to dry granulation. Thus, the slag particles are used to produce hot air or steam, becoming a source of thermal energy. In the cited methods, a very important issue is the grain distribution of slag particles, because the degree of waste heat recovery and gasification efficiency depend on this parameter. As in the case of granulation of other waste materials, the process parameters influencing the particle size distribution were examined, including rotational speed, liquid flow rate, liquid viscosity, and air flow rate [75,76]. Wang et al. used the computational fluid dynamics simulation method to visualize the centrifugal atomization process of molten slag particles on a spinning disk. Wang and colleagues also investigated the properties of continuous free-surface film and the particle size distribution of slag on and around the rotary disk [77,78]. Liu et al. investigated the granulation process of different types of metallurgical slags and heat recovery options depending on the rotational speed and particle size. For this purpose, a test stand was developed, consisting of a rotary cup with a diameter of 130 mm, rotating at a speed between 800 and 1500 revolutions per minute. The molten slag was poured through tundish onto the rotary cup, where it was broken into granules, which were then frozen and collected in this form by a circular collector. A high-speed camera was used to record the entire process, with the recording saved on a computer. The research showed that during granulation experiments of blast furnace and ferroalloy slag, the main mass fraction ranged from 2.44 mm to 3.14 mm. The mass fraction of granules with smaller diameters increased with increasing speed, but never exceeded that of the main fraction. The copper slag, in turn, showed a tendency to increase the fraction of small-diameter granules in the total weight of the granulate with increasing rotational speed. For the metallurgical slag, it was possible to find a tendency to homogeneity of the granules with increased rotational speed. Studies have also shown that in order to achieve high heat transfer in industrial applications, it is necessary to increase the rotational speed [79].

Summarizing, it can be stated that granulation processes are an effective method of recovering waste heat. In the next section, biochar granules will be discussed.

4.3. Biochar Granulates as a Solutions for Sustainable Economy

Biochar is formed in the pyrolysis process when the carbon dioxide that would be emitted into the atmosphere from biomass is bound in a solid form in an extremely stable form. Biochar is produced mainly from biomass residues generated by sawmill production, forestry, fruit and vegetable processing, and also agricultural production. Its source can be agricultural waste biomass such as corn stover, grass, rice husks, and straw, manure or poultry production, and pressing palm oil residues. It can also be produced from segregated biodegradable municipal waste or sewage sludge.

Biochar has a very high application potential. It could be used as biofuel, thus being an effective energy and heat source. Because of its chemical properties, it could also be used in agriculture as a soil amendment. It could also serve as an adsorbent of water or air pollutants in waste management [80]. In recent years, there has been much research on using biochar to improve soil quality and carbon sequestration [81–83]. This second application appears to have significant potential as a solution for mitigating climate change using biochar properties to block carbon dioxide. The difficult task, however, is to prevent the breakdown of biochar and its retention in the ground, thus avoiding releasing CO_2 into the atmosphere again. Biochar also has potential as an energy product based on or with the addition of various types of waste such as manure, segregated biodegradable municipal waste, or sewage sludge, and residues from the agriculture and wood industry [84,85].

The challenge in the effective use of biochar is its loose form, which makes it difficult to stay in the environment to which it is applied, and also poses a risk of irritation by coal dust. Therefore, the solution is to use granulation processes, which facilitate the use of biochar, e.g., as an additive to soil. Briens and Bowden-Green proved that the drum granulation process enables the effective formation of a given size of biochar granules. The research involved biochar from three plant biomass raw materials—corn stalks, birch bark, and miscanthus. All raw materials were successfully granulated; however, due to the differences in the hydrophobicity of the source material, they showed different characteristics of the granulation process. The granulated maize showed slightly hydrophobic properties, going from the phase of liquid marble formation, through collapsed liquid marble to coalescence and consolidation. Granulated birch bark showed moderately hydrophobic properties—the process started with the liquid marble formation phase, followed by coalescence, which led to the formation of large liquid marble formation, and then collapsed liquid marble, and in the final phase, again, coalescence and formation of the proper granules. Granulated miscanthus showed very hydrophobic properties, going from the partial marble phase, through liquid marble formation, followed by layering leading to the formation of the right granules. Granules from all three raw materials changed their properties depending on the process parameters. During granulation, increasing the rotational speed and the concentration of the binder additive affected the size and mechanical strength of obtained granules. The latter was high and further increased with speed increase and the volume of binder liquid, which also affected the granules' size expansion. The experiment clearly showed that drum granulation is applicable to various types of biochar, while an important issue is the selection of appropriate process parameters in order to obtain the final material with specific properties [86].

Due to their chemical properties, biochar and the ashes from biomass combustion processes are used in agriculture as fertilizer components. However, as already mentioned, the problem, due to their flowability, is transport and application to the soil. Therefore, granulation processes are used to obtain a homogeneous material with given properties. Vincevica-Gaile et al. conducted an experiment in which they granulated biochar with wood ash using freshwater sediment (sapropel) as a natural binder rich in organic substances. The purpose of the prepared granules was to use them as soil improvers. Studies have shown that the mechanical strength of granules obtained with drum granulation is higher than extruded granules. This process also increased the density of granules. The optimal mass ratio for granules was 30:100 for the mix of biochar with sapropel, while for the mix of fly ash with sapropel, it was 67:100. Obtained granulates had a size distribution from 3 mm to 8 mm and as research has shown, it can be used in agriculture or forestry [87].

The biochar granulation process can also be easily modified in order to obtain the appropriate properties of the granulate by using various additives and binding liquids. Researchers have performed granulation using different binders, i.e., HPMC, molasses, and NH_4NO_3. As a result of this experiment, it was found that hydroxypropyl methylcellulose as a binder showed moderately hydrophobic properties—the process started with the liquid marble formation phase, followed by coalescence, which led to the formation of large liquid marble formation and then collapsed liquid marble, and in the final phase, coalescence and formation of the proper granules. On the other hand, molasses and ammonium nitrate showed very hydrophobic properties—going from the phase of partial marble, liquid marble formation, and layering leading to the formation of the right granules. The obtained granulate was loose with size distribution ranging from 1 mm to 4 mm, depending on the rotational speed and the concentration and volume of the binder additive. The abrasion resistance, in turn, depended only on the latter parameter—the granules made with the addition of molasses had the highest resistance [88].

Another example of the use of biochar pellets as part of waste management is presented by Cairns et al. with their use for immobilizing certain heavy metals from highway runoffs. In their experiment, two types of granulates were used (<3 mm size). The first one was a mix of sintered larch biochar with wood ash (WASGr). The second one was a mix of larch biochar with wood ash cold (WAGr). Both types of granulate retain certain chemical elements, such as calcium, potassium, magnesium, sodium, and phosphorus. Those elements are necessary for the immobilization of heavy metals—lead, copper, zinc, and cadmium, better than wood ash. Of the two tested granulates, WASGr was more effective in immobilizing pollutants. Both granulates were rinsed with deionized water but this treatment did not have a negative effect on the high degree of immobilization of impurities, which ranged from 97% to 100%. This proves the effectiveness when applied to the immobilization of heavy metals in highway run-offs without any negative effects on freshwater ecosystems [89].

Biochar is often used in agriculture as a fertilizer ingredient or soil improver, though a team of researchers at the University of Toronto investigated its use as an additive to green roofs substrates in urban spaces. Green roofs are green design solutions increasing the sustainability of buildings in cities. They help mitigate the urban heat island effect by decreasing roof temperatures and thus cooling the buildings and the urbanization environment. Using biochar also helps improve air quality, collecting and filtering rain water, which contributes to cities' biodiversity. Biochar can be used in green roof solutions thanks to its unique properties, i.e., low weight, high nutritional properties, and water retention capacity. The drawback of biochar is that it is also vulnerable to wind and water erosion (because of loose form), which leads to a loss of positive effect on green roofs substrate. Scientists examined the use of granulated biochar in comparison to conventional biochar on drought-tolerant forb Agastache foeniculum. The results of the experiment proved that granulated biochar significantly improved plant growth and its reproduction, while at the same time neutralizing pH and collecting and filtering water. The best results were achieved using granules with 2–2.8 mm particle size. In conclusion, the use of agglomerated biochar is an effective method for increasing plant performance in green roofs solutions [90].

As previously mentioned, granulated biochar is considered as a perfect alternative to conventional soil additives. In recent years, because of its properties, there has been increased development of the biochar-based slow release of fertilizers (SRFs) [91]. One method aimed at improving the performance of biochar efficiency as a fertilizer is its granulation. This process also helps to overcome the high loss of conventional biochar particles due to wind and water while reducing its irritation to the eyes and respiratory systems of humans [92,93]. Granulation lowers the costs of transport and storage of biochar-based products [94]. Scientists are exploring various possibilities of using biochar as an SRF, for example, pelletizing biomass waste (manure compost) with biochar as an effective solution for NH_4^+-N adsorption, but also for increasing plant growth [95]. Another experiment used a drum granulator with a mix of wheat straw biochar, diammonium phosphate, potassium chloride, bentonite, and steam vapor as a binder [96]. The results showed that granulated fertilizer increased phosphor supply, moisture regime, grain yield, and maize production (accordingly by 10.7% and 46.2%) while also having a slow nutrient rate. Shi et al. obtained biochar SRF granulate composite by mechanically pelletizing a mix of biochar, bentonite, sepiolite, and urea with vinegar as a binder. The experiment showed that, compared to urea, the cumulative release of nitrogen and the dissolved organic carbon from the granulate decreased by more than 70% and 8%. Moreover, biochar granulate increased the maize shoot and root, by 14% and 25%, respectively [97].

The conducted review shows that granular biochar is an important alternative in the field of waste management towards biofuels, as well as its application in the agro sector as a soil improver and biostimulant for plants.

5. Conclusions

This work has shown granulation is an effective method of processing waste materials. Moreover, it has been shown that the properties of granules can be freely shaped depending on their intended use. In addition, the innovative aspect of the article is its focus on the broadest possible environmental aspect understood in minimizing the burden related to the amount and composition of waste generated by various industries.

The granulates presented in this review confirm that there is still untapped potential in pressure and non-pressure granulation processes. Moreover, various researchers and their works prove that granulation could provide effective solutions for various challenges in today's world, which are also in line with the goals of a sustainable economy as well as sustainable development. Granulates based on waste and/or bio-coal can be used as:

- A source of biofuel producing clean energy and thus reduce global emissions of carbon dioxide, which is in line with UN Sustainable Goal no. 7 and no. 13;
- Soil amendments or fertilizers improving agricultural production, which is in line with UN Sustainable Goal no. 2;
- Substrate amendments helping in improved collection and filtration of storm water, which is in line with UN Sustainable Goal no. 6;
- The source of processed bio and industrial waste that can be reused, as mentioned above, in numerous industries, which is in line with UN Sustainable Goal no. 12.

It is also worth noting that granulation processes are using easily accessible raw materials and additives, such as mineral fillers, water, etc., which means that the whole operation is relatively easy, waste-free, and cost-effective. Moreover, through variables such as the volume of resources used (fillers, binders) and process parameters (rotational speed, angle of inclination of the granulating apparatus), the properties of the obtained granulates can be easily selected depending on the demand and planned application. The number of possible granulation processes configurations is virtually unlimited.

The presented research is also in accordance with the principle of the European Green Deal, a set of policies focused on improving the quality of life for EU citizens by providing clean water, healthy soil and biodiversity, cleaner energy and technological innovations, and a resilient economy.

Recent advances show that there has been an intensification of research in the field of various agglomeration operations. However, more research still needs to be conducted in accordance with sustainable economic principles. For that reason, the authors of this paper will continue their innovative research in the area of optimizing waste and biomass granulation processes to develop, verify, and implement the model of the migration of waste components to the environment, with particular emphasis on waste from leather and tanning, agro food, and other industries.

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