Valorization of Bread Waste into Value-Added Products

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Bread is a universal food that is sold and consumed across the entire social and geographical spectrum. Bread waste is currently of increasing interest, as it is considered a huge global issue with serious environmental impacts and significant economic losses that have become even greater in the post-pandemic years due to an increase in cereal prices, which has led to higher production costs and bread prices. Meanwhile, many efforts have been initiated in the past decades to investigate methods of repurposing bread residues into fuel and chemicals such as bioethanol, biohydrogen, succinic acid, and various added-value products that can be exploited in versatile industries.

Keywords: bread residues ; valorization ; value-added products

1. Reprocessing Bread for Bread Making

One of the methods used in the baking industry to valorize low-quality bread and bread returned from the distribution network is reprocessing. The recycling of non-standard bread saves natural food resources and provides economic benefits for bakers.

Recycled bread offers many economic benefits for bread producers. Despite the low quality of recycled bread compared to the ingredients, which affect its luster, brittleness, softness, taste, specific aroma, and smell, the development of resource-saving technologies was found to be relevant. In addition, inappropriate storage conditions generate fungal mycelium on the bread surface; fungi enzymes break down proteins, fats, and carbohydrates, leading to a deterioration in the bread’s properties (unpleasant appearance, smell, and taste).

Savkina et al.[1] investigated the impact of recycled old bread on bread quality and its microbiological safety. Only fresh bread within the shelf-life date is allowed to be recycled in Russia. The authors studied the substitution of flour at 15, 20, 25, 35, 50, and 100% by recycling old breadcrumbs in sourdough samples. The obtained results showed that bread dosage in the rye-dense sourdough greater than 25% negatively affected the sourdough quality without any nutrient deficiency noticed. At a recycled bread dosage of 25%, the resulting quality was comparable to traditional rye-dense sourdough, but the crumbliness was 1.5 times lower compared to the control.

Sensory evaluation showed that bread made with fermented old bread in sourdough had interesting sensory characteristics (crust shape and color, taste, odor, chewiness, and porosity) comparable to the control. Moreover, old, recycled bread did not show any significant effect on the microbial contamination (molds and yeasts) of new bread.

The Holland Company Sonextra Sustain, a special starter for reprocessing bread, has developed a new concept for the reprocessing of bread. It offers to the customer the possibility to proceed safely to produce good-quality bread. This special technology included in the starter for reprocessing bread prevents the growth of bacteria that form spores and cause mold, resulting in tastier and softer bread.

Consequently, reprocessing bread is considered an alternative way to manage waste bread, as it contributes to tastier bread with upgraded quality, lower costs, and less environmental damage.

2. Ethanol Production

Ethanol is considered one of the most promising fuel sources. It may be used as an alternative fuel instead of fossil fuels. Sustainable bioethanol is mostly produced by microbial fermentation (commonly yeast) of agricultural and/or food wastes.

Among raw materials used in fuel ethanol production, starchy materials are the most common feedstocks. One of the most promising, highly accessible, and inexpensive raw materials is bread residue. It contains a significant amount of starch, a polymer composed of glucose molecules, which is easily hydrolyzed to monomeric sugars using amylases.

According to Dewettinck et al.[2], the amounts of starch and simple sugars in bread were found to be 500–750 and 3–50 g/kg, respectively. Moreover, bread waste contains 100–150 g/kg of proteins, which, after hydrolysis to peptides and amino acids, accelerate yeast growth and fermentation.

Pietrzak and Kawa-Rygielska [3] studied the direct conversion of starch in waste wheat–rye bread to ethanol using a granular starch hydrolyzing enzyme (GSHE). They suggested three raw material pretreatment methods, i.e., enzymatic prehydrolysis, microwave irradiation, and sonification, for ethanol yield and fermentation course improvement compared to separate hydrolysis and fermentation (SHF). The obtained results from the work conducted showed that all pretreatment methods increased the final ethanol yield compared to unpretreated waste bread fermentation. The fermentation of unpretreated waste bread ended with an 80.00% ethanol yield, while the use of pretreated raw material improved ethanol yield by 3–8%. Furthermore, the highest values of ethanol productivity were achieved in all studied samples within the first 48 h of fermentation.

Datta et al. detailed trials including the utilization of bread waste as the sole source to manufacture glucose using *Aspergillus niger* through solid-state fermentation, followed by the synthesis of bioethanol from glucose using *Saccharomyces cerevisiae* [4]. The solid-state fermentation of waste bread using *A. niger* produces a multienzyme solution containing amylolytic and proteolytic enzymes. The crude enzymatic extract was used for bread waste hydrolysis at 55 °C and 300 rpm, resulting in approximately 145 g/L of glucose. The hydrolysate was then used to produce ethanol at a concentration of 54 ± 2 g/L with an achieved conversion efficiency of 72%.

In the same context, Mihajlovski et al. [5] also optimized bioethanol production from bread waste as a biomass source by response surface methodology (RSM). The effect of fermentation duration (24–72 h) and waste brewer's yeast inoculum (1–4%) on ethanol production was studied. The optimized conditions, obtained by central composite design (CCD), were 48.6 h of fermentation and 2.85% of inoculum. Under these conditions, the maximum ethanol production of 2.06% was reached. The obtained results demonstrate that the use of waste bread offers multiple benefits related to environmental protection, reduction in production costs, and saving of fossil fuels.

Interestingly, a British beer company called "Toast Ale" have also used bread and bakery wastes to produce brews. In fact, beer is produced primarily from cereal grains (mostly barley), hops, and water. Recently, small breweries have started to use surplus bread in their recipes, substituting part of the malted barley, originally used as a source of sugar for fermentation. Two recipes were investigated in this research. In both recipes, 25–28% of the original malt was substituted with dried bread. The remaining ingredients were maintained as used in the standard process of beer production. These new beers are opening up a space in the craft beer industry, where the sustainability of food waste valorization options is needed in addition to flavor [6].

The described processes are important in the sustainable chemical industry because they convert waste food into value-added products such as ethanol. The previous methods of bioconversion of waste bread into alcoholic products limit the use of chemical additives, extra distillation, and purification steps and contribute to preserving the environment and human health along with lower production costs.

3. Lactic Acid Production

Lactic acid is one of the most important organic acids that has great value with variable applications in many industries such as food, beverage, pharmaceutical, chemical, and cosmetics. It has attracted the interest of many researchers, especially in the production of biodegradable poly lactic acid (PLA), well-known as a bioplastic material [7]. According to the report of Global View Research, the global lactic acid market size reached its highest peak in 2020, and it is expected to expand in the following years with an estimated annual growth rate of 0.8% [8].

The production of lactic acid can be performed by chemical synthesis or by fermentation using starchy or sugary biomass. Thus, the richness of bread waste with starch and other nutritional compounds provides a great renewable resource for lactic acid production in both processes.

The chemical conversion of bread residues into lactic acid can be carried out via hydrothermal treatment using alkaline catalysts (NaOH, KOH, $Ca(OH)_2$, LiOH, and K_2CO_3). As described in the study by Sánchez et al. [9], a mixture of bread and alkaline solutions in a ratio of 1/25 (solid/liquid) was heated at 300 °C with continuous stirring for 30 min. The maximum yields of lactic acid production of 38.11 ± 0.2%, 34.46 ± 0.21%, and 72.90 ± 4.45% were reached in the mixture of KOH (0.4 M), NaOH (0.6 M), and $Ca(OH)_2$ (3.5 M), respectively.

Despite the time-saving advantage presented by the previous method, it was not able to effectively eliminate the environmental side effects of the production of undesirable and harmful compounds, i.e., lactic acid D-isomer [7]. Hence, researchers tend to lean towards the fermentation process due to environmental impact and full control of the product outcome. In other words, an optically pure L(+)- or D(−)-lactic acid can be obtained by microbial fermentation of renewable resources depending on the microorganism used [10]. The fermentative production of lactic acid is based on the conversion of sugary, starchy, or even lignocellulosic biomass via different strains of microorganisms. The fermentation process also has its challenges. The yield of fermentation can be significantly affected by various factors, namely, physiochemical factors (pH, temperature, nutrients, substrate concentrations, etc.), the microorganisms, and the type of biomass used [11].

Furthermore, Pleissner et al. [12] experimentally investigated the fermentation method using restaurant food waste as starchy feedstuff. This process was based on simultaneous saccharification and fermentation through two different strains (*Streptococcus* sp. and *Lactobacillus* sp.). Maximum productivity of 2.16 g L^{-1} h^{-1} was observed in *Streptococcus* sp. strain, yielding 58 g/L of lactic acid from a 20% (*w*/*w*) food waste blend.

Recently, Hassan et al. [13] introduced another process of fermentation using a combination of kitchen food waste and banana peels in a ratio of 3/1 (*w*/*w*) using *Enterococcus durans* BP130. Although this process was conducted under harsh conditions, it delivered interesting results: maximum productivity of 0.24 g L^{-1} h^{-1} yielding the highest lactic acid concentration of 28.8 g/L.

4. Succinic Acid Production

Succinic acid (SA, $C_4H_6O_4$) is a dicarboxylic acid that has attracted great interest worldwide. The presence of two carboxyl groups makes SA a precursor molecule for the synthesis of many chemical compounds. Hence, it has a wide range of applications in industries such as pharmaceuticals, food, polymers, plasticizers, and green solvents [14].

Originally, succinic acid was produced via a fossil-based system through catalytic hydrogenation of petrochemical maleic anhydride. However, this process can be expensive and harmful to the environment [15].

Another route for SA synthesis is bio-based production via fermentation from renewable resources. This process has attracted research interest because it provides lower energy consumption due to milder operation conditions and lower dependence on a single feedstock. The common microorganisms used in fermentative SA bio-production are *Actinobacillus succinogenes*, *Anaerobiospirillum succiniciproducens*, *Mannheimia succiniciproducens,* and recombinant *Escherichia coli* [16]. Most of these microorganisms use glucose as a carbon source for SA production. Thus, due to its richness of carbohydrates and other nutrients, bread waste was found to be an excellent substrate for SA biosynthesis.

In general, the fermentation process is carried out through two main steps. Firstly, the processing of bread waste, via hydrolysis using solid-state fermentations of *Aspergillus awamori* and *Aspergillus oryzae,* produces enzyme complexes rich in amylolytic and proteolytic enzymes, respectively. As described in the study of Leung et al. [17], this step generates a hydrolysate containing over 100 g/L glucose and 490 mg/L free amino nitrogen. Moreover, the bread hydrolysate is then used as the sole feedstock for *Actinobacillus succinogenes* fermentation, leading to the production of 47.3 g/L succinic acid with a yield and productivity of 1.16 g SA/g glucose and 1.12 g/L h, respectively. Overall, 0.55 g of succinic acid per gram of bread was obtained via this process [17]. The biochemical pathway for SA production from glucose (carbon source) by *A. succinogenes* is described by Gadkari et al. [14] as follows:

PEP/Pyruvate carboxylase route: $0.5\ C_6H_{12}O_6 + NADH + H^+ + CO_2 \rightarrow C_4H_6O_4 + H_2O + NAD^+$

PEP carboxy kinase route: $0.5\ C_6H_{12}O_6 + CO_2 + NADH + H^+ + ADP + Pi \rightarrow C_4H_6O_4 + H_2O + NAD^+ + ATP$

Following these experimental demonstrations, Zhang et al. [18] conducted the same fermentation process by using cake and pastry wastes separately as feedstocks, producing SA at 24.8 and 31.7 g/L with a yield of 0.80 and 0.67 g SA/g sugar, respectively. According to this research, the SA productivity achieved with cake and pastry wastes was 0.79 and 0.87 g L^{-1} h^{-1}, with an overall yield of 0.28 and 0.35 g succinic acid per gram of substrate, respectively.

5. Biohydrogen Production

Hydrogen is considered a valuable component due to its possible use as a promising energy source in the future because it is clean and renewable. Hence, many studies were conducted to find the best production process, such as steam reforming, electrolysis, biophotolysis of water, and fermentation. Most researchers seem to prefer the fermentation technique for H_2 generation because it is eco-friendly and requires less external energy [19].

Bread waste is an excellent biomass for biohydrogen (H_2) production due to its wide range of nutrients.

The process of biohydrogen production can be divided into two steps. Firstly, the processing of bread waste biomass enables the breakdown of all nutrients as macromolecules (starch and protein) into monomers (glucose and free amino nitrogen). As mentioned in the research of Han et al. [20], the hydrolysis of this biomass can be performed via crude enzymes, which were generated by two microorganisms: *Aspergillus awamori* and *Aspergillus oryzae* via solid-state fermentation. In the second step, the waste bread hydrolysate was then used for biohydrogen production by anaerobic sludge in a continuously stirred tank reactor (CSTR). This was the first research that reported continuous biohydrogen production from waste bread by anaerobic sludge with a yield of 109.5 mL hydrogen/g of waste bread at chemical oxygen demand (COD) concentration of 6000 mg/L [20].

Recent research has thoroughly investigated other processes such as dark fermentation using a wide range of substrates including agricultural and industrial starchy wastes with a low level of undesirable compounds. This process was theoretically shown in the past to be a fruitful process for hydrogen production with mild operation [21].

Recently, Jung et al. [22] showed the feasibility of dark fermentation for biohydrogen production. They stated that conducting the process of dark fermentation from food waste using hybrid immobilization in mesophilic conditions yielded a hydrogen production rate (HPR) of 9.82 ± 0.30 L/L-d at an organic loading rate (OLR) of 74.7 g hexose/L-d, leading to a yield of 1.25 ± 0.04 mol H_2/mol $\text{hexose}_{\text{consumed}}$ [22]. However, this framework solely pertains to a laboratory scale, and the scaling-up option is yet to be tested.

6. Hydroxymethylfurfural (HMF) Synthesis

Bread waste offers renewable biomass that can be exploited in hydroxymethylfurfural (HMF) production. Also known as 5-(hydroxymethyl)-2-furancarboxaldehyde and 5-(hydroxymethyl)-2-furaldehyde, HMF displays a wide range of applications in versatile industries due to its capacity to be upgraded into many chemicals such as medicines, polymers, resins, fungicides, and biofuels [23].

Fundamentally, the synthesis of HMF from food waste involves three reactions: hydrolysis of glucan to glucose, isomerization of glucose to fructose, and dehydration of fructose to HMF.

Previous research by Yu et al. [24] showed that HMF production from bread waste is based on the thermochemical process using a mixture of polar aprotic solvents (DMSO, THF, ACN, or acetone) and water in a ratio of 1/1 (*v*/*v*) under heating at 140 °C with $SnCl_4$ as a catalyst. Under these conditions, a maximum HMF yield of 26–27 mol% was achieved in the mixtures of ACN/H_2O, acetone/H_2O, and DMSO/H_2O. However, this method fails to eliminate any undesirable side reactions (rehydration of HMF to levulinic acid and polymerization of HMF to humins), which can significantly affect the production yield [24].

The applicability of these results was then tested in the research of Cao et al. [25][26] on starchy food waste (i.e., bread, rice, and spaghetti), showing a yield much higher than the previous study. They began by testing two different biochars as a catalyst followed by optimizing the reaction conditions to heating at 180 °C for 20 min in a mixture of dimethylsulfoxide (DMSO)/deionized water (DW) in a ratio of 3:1 (*v*/*v*). Under these optimum conditions and by using sulfonated and acid-activated wood biochars, the yield of the conversion was found to be improved to reach 30.4 Cmol% [25] and 30.2 Cmol% [26] respectively. This study expanded the understanding of the effects of biochar catalysts on desirable and undesirable reactions, showing higher catalytic activity of biochars towards starch hydrolysis and fructose dehydration.

7. Bread Waste for Protein and Pigment Production

One of the main value-added products of waste bread is proteins and pigments, such as carotenoids, which are present in photosynthetic microorganisms and plants. They are tetraterpene pigments that display orange, yellow, red, and purple colors [27]. These isoprenoid pigments are used as nutraceuticals and health additives and have been recently developed to be fit for pharmaceutical uses to prevent many diseases, including cancer. They are known for their antioxidant characteristics, which confer the ability to protect cells against oxidative damage by neutralizing free radicals [28].

Carotenoids are also used as natural pigments in the food industry as additives to conserve the shelf life of many food products or to enhance organoleptic features [29] and are used in many other industrial areas as well, such as paper and textile industries.

Gmoser et al. [30] used waste bread as a substrate for the edible filamentous fungus *Neurospora intermedia* in a double-staged fermentation process combining submerged and solid-state fermentations to produce pigments such as carotenoids. Submerged fermentation was performed initially on thin stillage to produce the fungal biomass *N. Intermedia.* The latter was then used for solid-state fermentation in the presence of air on waste bread to generate a feed rich in carotenoids and proteins. The obtained results showed that the production of carotenoids was noticeably higher than that obtained using inoculation with a spore solution. Furthermore, proteins showed an increase of approximately 161% compared to the bread waste before the fermentation process.

Another study conducted by Haque et al. [31] investigated the production of natural pigments (orange, yellow, and red) applied in food and textile industries. The authors used recovered amino acids and sugars from bakery waste hydrolysate as substrates for the filamentous fungus *Monascus purpureus*, as well as bakery waste, to produce proteins (enzymes) such as glucoamylase and protease. The bakery waste hydrolysate was generated using different species of filamentous fungi *Aspergillus awamori* and *Aspergillus oryzae* and was then used as a substrate for *M. purpureus* in submerged fermentation. The same fungus that used bakery waste as a substrate in solid-state fermentation was investigated to produce the enzymes glucoamylase and protease. The highest yield of pigment, which presented around 24 AU units/g glucose, was obtained with the bakery hydrolysate with a low initial glucose concentration of 5 g/L.

Moreover, glucoamylase and protease enzyme activities were 8 U/g at 55% of moisture and 117 U/g at 60% of moisture, respectively, at 30 °C [31].

8. Bioconversion of Waste Bread to Glucose–Fructose Syrup

Many have studied green and sustainable methods to convert waste bread into fermentable sugars, as it is mainly composed of starch (natural polymer consisting of polysaccharides).

In fact, the bioconversion of waste bread into glucose–fructose syrup was investigated by Riaukaite et al. [32], involving enzymatic hydrolysis to produce glucose via a two-step process using amylolytic enzymes and then isomerization to produce fructose.

- The first step of the enzymatic hydrolysis process is the liquefaction of bread slurry to produce oligosaccharides, such as dextrin, using an α-amylase enzyme that breaks down long chains of polysaccharides making up the starch into shorter chains [33].

- The second step of this process is using the glucoamylase enzyme to break down dextrin into monosaccharides—actual glucose molecules [34].

Then, fructose is produced by enzymatic isomerization of the glucose molecules using the glucose isomerase enzyme. High yields of the final glucose–fructose syrup were found to depend on the amount of bread and enzymes used [32]. In fact, bioconversion of a minimal amount of waste bread resulted in low yields of glucose. Additionally, a high amount of waste bread induced a highly viscous slurry, halting enzyme activity in breaking down starch [35]. According to Riaukaite et al. [32], the produced glucose–fructose syrup was composed of 45.27 ± 0.55% glucose and 40.32 ± 0.80% fructose.

Another study conducted by Haque et al. [36] investigated the bioconversion of beverage waste to fructose syrup as a low-cost value-added product in a multi-step process that involves enzymatic hydrolysis, activated carbon treatment, ion-exchange chromatography, and ligand-exchange chromatography. As a result, 47.5% of sugars present in the hydrolysate were recovered as a syrup rich in fructose, while the remaining sugars were in the glucose-rich stream.

9. Production of Aroma Compounds

An aroma compound, also known as an odorant, fragrance, or flavor, is a chemical compound characterized by a smell or odor when two conditions are established. On one hand, the compound must be volatile. This helps it to easily be detected by the olfactory system in the upper part of the nose. On the other hand, a sufficiently high concentration is an important factor to be able to interact with one or more of the olfactory receptors [37].

Aroma compounds constitute one of the most important segments in the perfumery, food, and cosmetic industries. Hence, the global flavors and fragrances market size reached a high peak in 2019 with an estimated scale of USD 28,193.1

million, and it is projected to reach USD 35,914.3 million by 2027 with an estimated compound annual growth rate (CAGR) of 4.7% [38].

In general, the production of aroma compounds can be carried out via chemical synthesis, extracted directly from a natural matrix or derived from biotechnological processes [39]. However, research leans towards the fermentation process using microorganisms because it is an economic alternative to the difficult and expensive extraction from raw materials such as plants and avoids the environmental impact of the chemical process [40], in addition to consumer preferences for products labeled as "natural" [41].

Studies have shown that solid-state fermentation is a promising process for aroma compound production. Due to its capacity to provide a complex aroma profile, bread waste can be used as an excellent feedstock for the fermentation process without the need for extra nutrients [42][43].

In previous research, Daigle et al. used *Geotrichum candidum* ATCC 62,217 to study the production of fruity fragrance compounds (pineapple-like) on fermented waste bread (35% white breadcrumb and 65% water) [40]. Because of this strain's affinity for aromatic compounds, it was able to convert the organic acids in bread into fatty acids esters, primarily ethyl esters of acetic, propionic, butyric, and isobutyric acids. The fermentation temperature, agitation, and time were only a few of the parameters that the authors optimized. Thus, after 48 h at 30 °C, the produced volatile chemicals reached their optimal levels.

Terpenes, the largest class of naturally occurring aromatic hydrocarbons that serve as a foundation for smell and flavor, can also be produced from bread waste. This strategy was examined in a recent work by Styles et al. [44] who used thermophilic bacteria in a metabolic engineering project. In this work, bread waste was employed as a feedstock for *Parageobacillus thermoglucosidasius* NCIMB 11,955 due to its high starch content (80%). This strain can directly break down starch to liberate maltose, which can be catabolized as a carbon source, characterized by starch-degrading enzymes in its genome, including -amylase and neopullulanase. After 48 h of growth and at a temperature of 55 °C, the highest production yield of terpenes was noted in a 2% bread waste media (*w*/*v*) versus maltose media (2% (*w*/*v*)). However, this production pertains to a laboratory scale and needs further experiments to convert it into industrial production [44].

Prior research has thoroughly investigated other food waste for aroma compound synthesis. Aggelopoulos et al. [42] appraised the formation of high amounts of Ɛ-pinene (which has chemical and physical properties similar to α-pinene and β-pinene) using food waste mixtures (orange and potato pulp) via solid-state fermentation. For this fermentation process, they used a natural mixed dairy culture consisting of symbiotic consortia of yeasts and bacteria embedded in a polysaccharide matrix (Kafir), which yielded an estimated production rate of 4 kg/ton of the treated substrate.

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