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Review

Agro-Industrial Food Waste as a Low-Cost Substrate for Sustainable Production of Industrial Enzymes: A Critical Review

Vishal Sharma [1,2,†], Mei-Ling Tsai [2,†], Parushi Nargotra [2,†], Chiu-Wen Chen [1,3], Chia-Hung Kuo [2], Pei-Pei Sun [2,*] and Cheng-Di Dong [1,3,*]

1 Department of Marine Environmental Engineering, National Kaohsiung University of Science and Technology, Kaohsiung 81157, Taiwan
2 Department of Seafood Science, National Kaohsiung University of Science and Technology, Kaohsiung 81157, Taiwan
3 Institute of Aquatic Science and Technology, National Kaohsiung University of Science and Technology, Kaohsiung 81157, Taiwan
* Correspondence: ppsun@nkust.edu.tw (P.-P.S.); cddong@nkust.edu.tw (C.-D.D.)
† These authors contributed equally to this work.

Abstract: The grave environmental, social, and economic concerns over the unprecedented exploitation of non-renewable energy resources have drawn the attention of policy makers and research organizations towards the sustainable use of agro-industrial food and crop wastes. Enzymes are versatile biocatalysts with immense potential to transform the food industry and lignocellulosic biorefineries. Microbial enzymes offer cleaner and greener solutions to produce fine chemicals and compounds. The production of industrially important enzymes from abundantly present agro-industrial food waste offers economic solutions for the commercial production of value-added chemicals. The recent developments in biocatalytic systems are designed to either increase the catalytic capability of the commercial enzymes or create new enzymes with distinctive properties. The limitations of low catalytic efficiency and enzyme denaturation in ambient conditions can be mitigated by employing diverse and inexpensive immobilization carriers, such as agro-food based materials, biopolymers, and nanomaterials. Moreover, revolutionary protein engineering tools help in designing and constructing tailored enzymes with improved substrate specificity, catalytic activity, stability, and reaction product inhibition. This review discusses the recent developments in the production of essential industrial enzymes from agro-industrial food trash and the application of low-cost immobilization and enzyme engineering approaches for sustainable development.

Keywords: enzymes; agro-industrial food waste; biocatalysis; immobilization; enzyme engineering

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1. Introduction

As global concern over food and agricultural sustainability, environmental resilience, and food safety has grown over the years, the food industry has been exploring more environmentally friendly ways to produce food and nutritional supplements. Enzyme biocatalysis, which operates at the nexus of microbiology, molecular biology, biochemistry, and organic chemistry is an illustration of a sustainable multidisciplinary technology. Biocatalysis has gained enormous industrial potential owing to its influential and eco-friendly prospects with effective kinetics and commercial benefits [1]. Enzymes are macromolecular biocatalysts with extensive applications due to their ability to operate in milder reaction conditions, high catalytic efficiency, superior product selectivity, and their negligible toxicity to the environment and the body [2,3]. They have been the subject of numerous research projects around the world in order to generate novel significant industrial processes. A variety of microbial species (bacteria, fungi) have been traditionally employed in the production of diverse products of commercial importance from different organic

substrates converting them into simpler forms through enzymes [4]. Many microbes that are typically utilized as a source of enzymes have had their microbial endogenous and exogenous enzymes thoroughly researched [5]. A significant portion of microbial enzymes are utilized in a variety of industrial processes, including those for food processing, animal feed, biofuels, paper and pulp industries, pharmaceutical industries, textiles, polymer synthesis, and detergent industries [6]. Enzymes such as cellulases, xylanases, amylases, lipases, proteases, and pectinases have been commercially utilized in a wide range of industrial processes, especially in the food industry.

In recent times, the idea of 'circular economy', which refers to the use of organic waste from one industry as a source of raw materials for another, has gained much popularity [7]. It is based on the principle of sustainable development known as the '5Rs' (reduce, recycle, reuse, recovery, and restore) and replaces the traditional linear economic model (make–use–throw) with a more efficient circular one [8]. The food and agro-industrial sectors have been revolutionized owing to modernization and industrialization, which has dramatically increased the production of huge amounts of agro-industrial food waste [9]. The United Nations' Food and Agriculture Organization (FAO) estimates that every year, about 1.3 billion tons of food, which is one-third of global production, is wasted [10]. In addition to the food waste, various agro-industrial residues and crop waste in the form of lignocellulosic biomass (LCB) is generated annually around the globe [11–16]. Most of this plant-based waste is either landfilled or burned alongside other municipal combustible trash in an effort to recover energy [17]. Apparently, this organic refuse, which is a rich source of carbohydrates, proteins, lipids, organic acids, and other necessary minerals, can be channelized towards its value addition [9,18]. It could serve as an inexpensive fermentation source for microbes in the food industries, which digest it via enzymes into key components of circular economies. The industrial applications of enzymes have significantly risen in the last decade, primarily in the food modification, biofuels production, biomedical and pharmaceutical research, and the transformation of agro-industrial waste [18–20].

Even though enzymes offer many more benefits over traditional chemical catalysts to valorize organic waste, a major bottleneck in their commercial viability is their non-reusability, high sensitivity, and poor catalytic activity and stability in extreme environmental conditions of temperature and pH [18]. These challenges therefore need to be critically removed through the development of stable biocatalytic systems. Enzyme immobilization has received significant attention in the past few years as an important engineering approach to customize and enhance a wide range of catalytic features of enzymes, including their activity, specificity, selectivity, and tolerance to inhibitors [12,19]. The development of flexible carriers, including agro-food and crop-based materials, metal organic frameworks, and nanomaterials, allows for the cost-economic immobilization of enzymes with better enzymological properties, enabling catalytic reactions to be carried out under rigorous processing environments [21].

The production of engineered enzymes by promising protein engineering tools such as directed evolution, rational design, and computational methods, aids in improving the enzymological properties with increased purity, catalytic efficiency, specificity, and expression yield, owing to the altered amino acid sequence [22]. The application of tailored enzymes for food processing enables their cost-effective production to achieve sustainable development. Figure 1 shows schematic representation of enhancing the value of enzymes by immobilization and protein engineering approaches through valorization of agro-industrial food/crop waste.

This review encompasses the valorization possibilities of agro-industrial food waste through microbial enzymes. The review also highlights the novel strategies for food enzyme immobilization and their potential applications in the food industry. Moreover, the deeper insights on development of engineered enzymes for sustainable and green processing of the waste biomass into diverse bioproducts are highlighted.

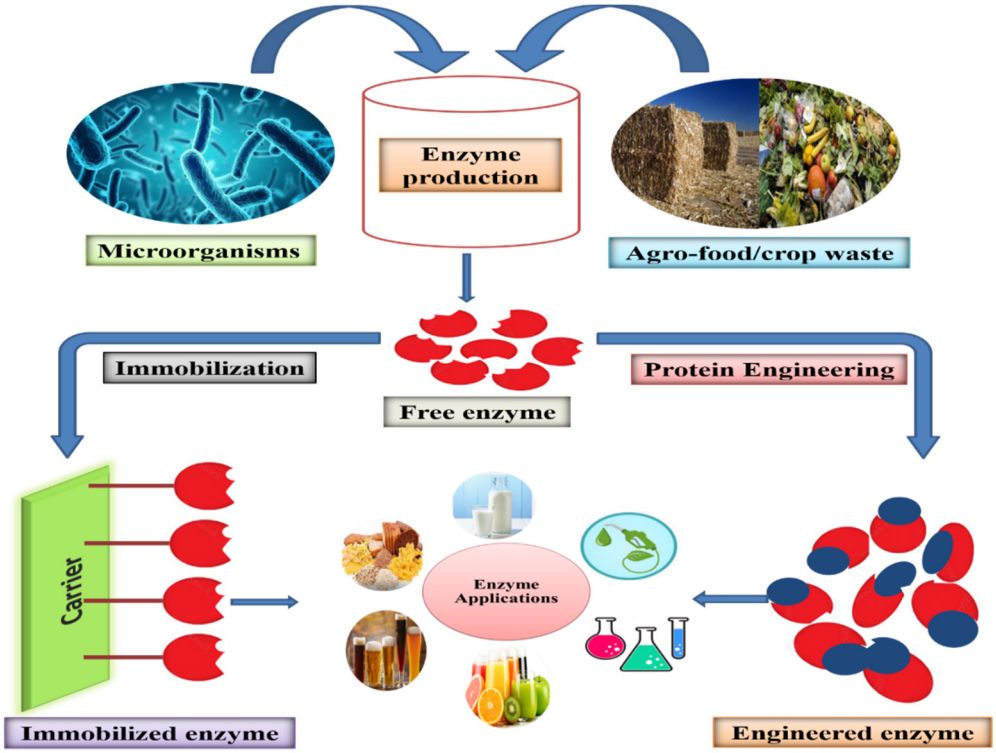

Figure 1. Schematic representation of developing efficient biocatalysts.

2. Types of Agro-Industrial Food Waste

The ever-increasing world population and the rapid urbanization and lifestyle changes have overburdened the food processing industries that generate abundant amounts of plant, animal, and agricultural residue wastes. The massive build-up of such natural food waste is a big challenge for mankind due to the lack of efficient waste management techniques. Commonly, within the food supply chain, food which is abandoned, unused, thrown out, and burnt from the harvest source or slaughter source is referred to as food loss. Most of the food waste is generated during its production, harvesting, transportation, industrial processing, and consumption [7]. The different types of organic wastes are majorly derived as agricultural wastes or those from the food processing industries. The agricultural waste can be majorly categorized as crop residues after harvesting, agro-industrial wastes, and fruit and vegetable waste [4,23], whereas the food processing industries generate enormous amounts of waste from cereals and pulses, fruits and vegetables, dairy, poultry, meat, egg waste, aquatic life waste, and seafood waste during their processing. Moreover, commercial and household kitchen waste also adds to the overall concerns of waste management [24]. Therefore, the apprehensions of deteriorating environment and waste management can be better overcome by transforming the agro-food waste into a substrate for enzyme production, besides a raw material for generation of diverse bioproducts such as biofuels, xylitol, xylooligosaccharides, bioplastics, organic acids, etc. (Figure 2). Emphasis must be laid on implementing modern technologies as well as alternative strategies for the effective and successful use of non-consumable portions from the agro-food wastes leading to proper waste management and sustainable development.

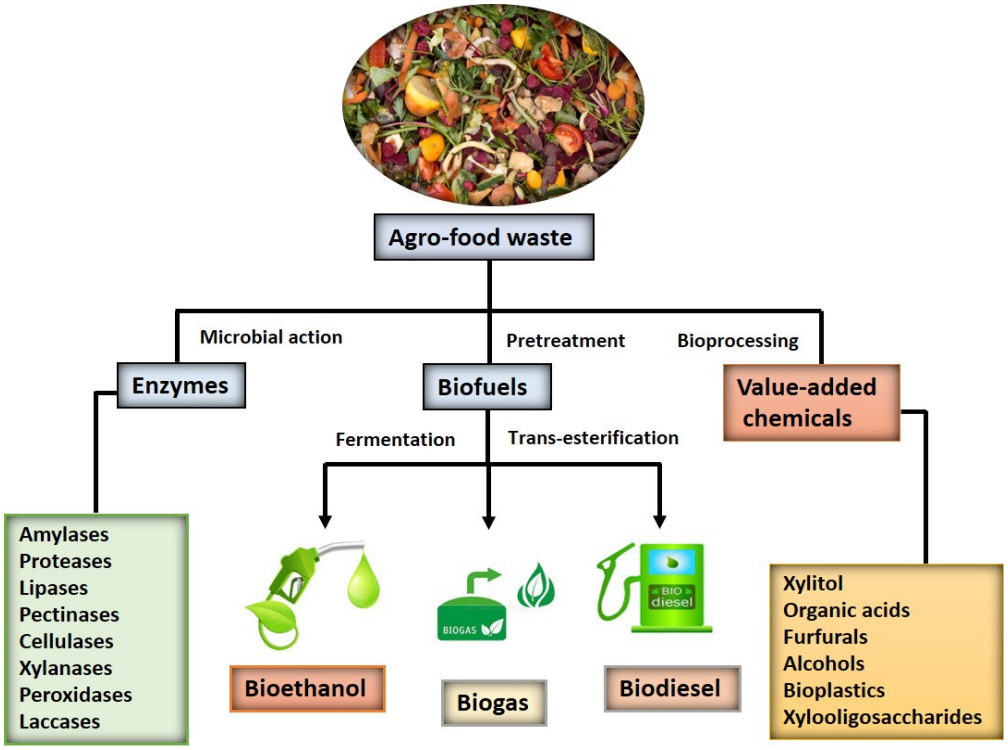

Figure 2. Agro-industrial food waste as a raw material for the production of different bioproducts.

2.1. Food Processing Waste

Different food processing and packaging industries produce tons of food items to meet the constant demands of the food market. However, the side generation of unprocessed food, leftover materials, spoilage, or contamination of raw food materials is an unavoidable practice in the food industries. The food waste is subcategorized majorly as starch-based food waste, dairy waste, meat and fish waste, vegetable trimmings, fruit peels, spent grains, and pulp trash. Traditionally, the leftover food from the food processing industries is dumped, incinerated, landfilled, composted, or anaerobically digested to enrich the soil. These food wastes, however, are extremely nutritious, which makes them substrates for microbial growth and production of diverse enzymes [25]. Moreover, these wastes can be used as inexpensive raw materials for the production of primary and secondary metabolites using microorganisms, with commercial value.

2.1.1. Fruit and Vegetable Waste

Fruit and vegetable waste (FVW) is produced in the food processing industries as a result of cutting, thermal treatment, handling, processing, packaging, transportation, and natural ripening [26]. Additionally, wastes are generated by microbial attack, discoloration, and by a number of biochemical reactions involving enzymes, antioxidants, phenolic chemicals, and oxidation [4]. The landfilling and incineration of food waste is a common practice preferred by industries, despite being unaffordable due to emissions of greenhouse gases and high capital and operating costs [17]. The anaerobic digestion of FVW into biofertilizers is an alternate option that might indirectly minimize environmental pollution and further increase soil nutrition. In many developing nations, FVW consisting of pineapple, papaya, banana leaves, orange peels, spinach, sugarcane tops, cabbage, etc., are converted into animal feed to address the current animal feed scarcity [8,9,27].

2.1.2. Edible Oil Waste

Cooking or edible oil waste is produced by the edible oil industries throughout a number of processing phases, including neutralization, degumming, deodorization, bleaching, and hydrolytic/oxidative rancidity. The hydrolytic rancidity of the waste

cooking oil results from lipid oxidation, moisture, age, oxidation, and addition of effluents from industry rich in carbohydrates, fatty acids, and proteins [28]. Waste cooking oil is a left over, dark colored liquid formed by repeated deep-frying processes, which is unfit for human consumption because of its free short-chain fatty acids, aldehydes, ketones, mono/di-glycerides, aromatic compounds, polymers, and many other properties [26]. Edible oil industrial waste is a cheap substrate for microbial lipid production that can be produced by its co-fermentation with food waste [29].

2.1.3. Kitchen Waste

With a rapid increase in population, urbanization, and economic development over the years, an abundant amount of kitchen waste (KW) is generated every day from households, restaurants, public catering rooms, and hotels. KW includes cooked food wastes, leftover fruits and vegetables, meat, shells and pits, egg shells, used oil, and grease [26]. It is typically thought of as organic waste that decomposes quickly and has unpleasant aromas that attract insects and rodents. KW residues are rich in carbohydrates, proteins, lipids, lignin, organic acids, inorganic salts, and other bioactive substances and can therefore be attractive substrates for enzyme production by microorganisms [30]. Humidified bread waste was used as a low-cost substrate for the coproduction of α-amylase and protease through solid state fermentation (SSF) by *Rhizopus oryzae* [31]. The utilization of KW with the goal of producing value-added goods by enzymatic reactions has the potential to improve the food supply chain and, therefore, food security.

2.1.4. Poultry, Slaughterhouse and Egg Processing Waste

A significant amount of waste is generated from the poultry, egg, meat, and related food processing industries, in addition to solid/liquid waste from slaughterhouses. A considerable amount of chicken feather waste is produced by the poultry industry, which could be an excellent source of protein (∼90% keratin) for various industrial applications [32]. Bacteria such as *Bacillus* sp. FPF-1, *Brevibacillus* sp., *Chryseobacterium* sp. FPF-8, and Nnolim-K2 were isolated as showing an excellent keratinase-producing ability employing chicken feathers as the substrate [33]. The waste from slaughterhouses includes skin, hairs, feathers, hooves, horns, deboning residues, and other materials that have a high level of organic matter, protein, and animal fat [4,34]. Slaughterhouse waste might pose major health and environmental risks if left untreated, which can instead be easily applied as a raw material in the production of a variety of commercial materials.

2.2. Agricultural Waste

Agricultural waste (AW) is produced in millions of tons annually and its improper management and disposal pose negative effects on the environment, including damaging the ecosystem. Therefore, transformation of AW into valuable products using economical, environmentally friendly, and sustainable methods has been the persistent objective of governments, environmentalists, and other stakeholders. AW, when properly managed and utilized, has the potential to be a significant contributor to ecological sustainability and energy security. Generally, AW in the form of bagasse, bran, husks, peels, leaves, seeds, stems, stalks, etc. (Figure 3) are utilized as soil improvers, fertilizers, animal fodder, and in various other processes [4,23]. Agro-waste as lignocellulosic biomass (LCB) is majorly constituted by cellulose backbone, with hemicellulose and lignin as other vital carbonaceous fractions [13]. These polymers from the LCB have been researched as potential substrates for lignocellulosic biorefineries for the production of second-generation biofuels and other valuable materials [35–39]. The enzymatic saccharification of complex polysaccharides into simple fermentable sugars using cellulolytic enzymes like cellulases and xylanases is an inevitable step in the systematic conversion of LCB into biofuels [37,38]. Solid-state fermentation (SSF) is also an attractive method for the exploitation of agricultural residues for production of a consortium of industrial enzymes.

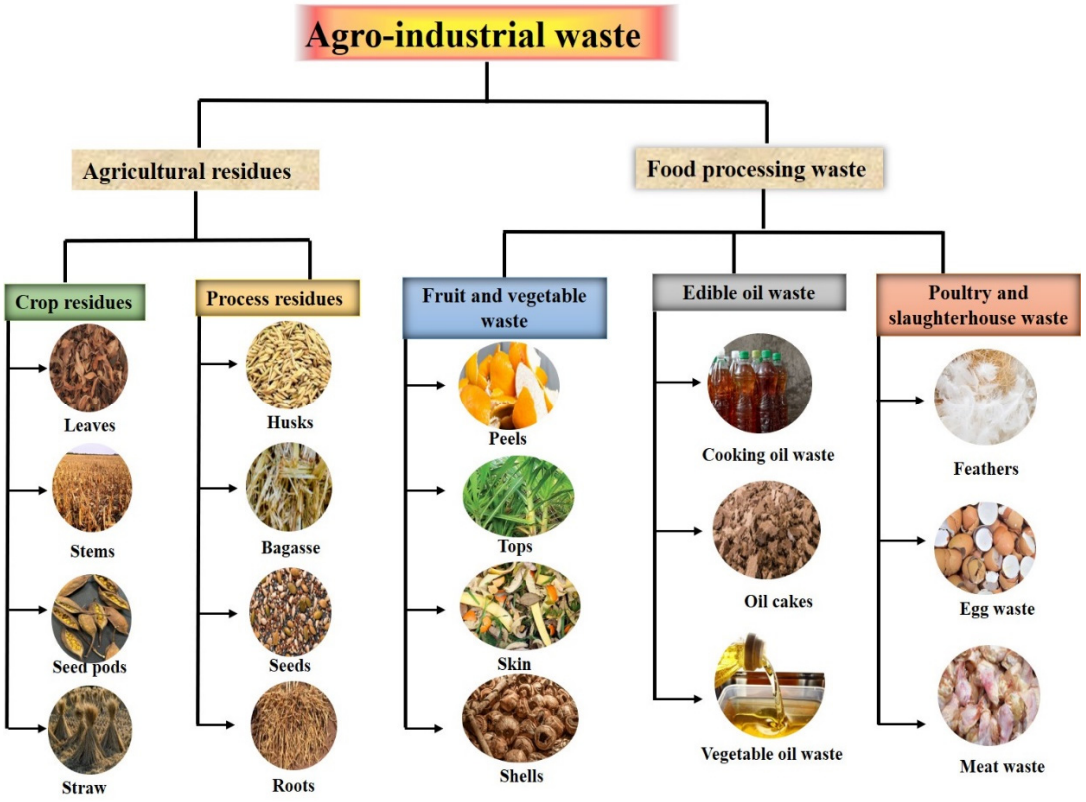

Figure 3. Classification of different types of agro-industrial food waste.

3. Production of Microbial Enzymes from Agro-Food Wastes

Traditionally, the microbial enzymes have been of much importance in food preparation techniques. The state-of-the-art developments in enzyme technology over the last few years have led to the creation of novel enzymes with a broad range of applications in several industries. These industries are majorly associated with biofuel production, food modification, agro-industrial waste transformation, laundry, and pharmaceutical and biomedical research [18]. Microorganisms including fungi, yeast, and bacteria, as well as their enzymes, are frequently utilized in a variety of food preparations to enhance flavor and texture, and they also provide enormous economic advantages to various enterprises [40]. The majority of the world's enzyme application is categorized into two categories as special enzymes used in research, therapeutics and diagnostics, and industrial enzymes for food and animal feed industries. The market for these enzymes is estimated to reach at about $7 billion USD by the year 2023 and is expected to increase at a 7.1% compound annual growth rate from 2020 to 2027 [41,42]. The valorization of agro-industrial food wastes by the production of low-cost enzymes under solid-state fermentation is a promising and extensively explored method [17]. Different sets of enzymes such as amylases, proteases, lipases, laccases, cellulases, xylanases, and pectinases, among other enzymes, have been produced by SSF using inexpensive food wastes. SSF offers a number of benefits, including lower cost, higher yield, less waste, and simpler equipment and culture media derived from organic, solid agricultural products or waste.

3.1. Amylases

Amylases are one of the most important industrial enzymes divided into two major subclasses, i.e., α-amylase (EC 3.2.1.1) and glucoamylase (EC 3.2.1.3). α-amylase cleaves 1,4-α-D-glucosidic linkages in starch to convert it into maltose, glucose, and maltotriose, whereas glucoamylase specifically converts amylose and amylopectin's non-reducing ends to glucose [43]. A-amylases, however, have been widely explored and applied in food, clinical, brewing, detergent, textiles, and paper industries [17,44]. Additionally, they have

also been widely explored for the valorization of agro-industrial residues and organic by-products to improve the generation of bioproducts. Different microbial strains may produce high activity amylases under optimized fermentation conditions using agro-food wastes, such as kitchen waste, potato peels, watermelon rinds, and crop residues such as rice husks [45] and corn cobs, coffee waste, and tomato pomace. Iqbalsyah et al. [45] investigated the production of amylase using rice husk substrate via solid-state fermentation by *Geobacillus* sp. with maximum amylase activity of 1.85 U/g at 48 h. Mojumdar and Deka [43] produced α-amylase by *Bacillus amyloliquefaciens* in SSF using rice bran, wheat bran, and potato peels as common agro-industrial feedstocks. The medium containing wheat bran as a substrate yielded the highest titer of amylase activity (112 U/mL), which was followed by that of potato peels (89 U/mL) and rice bran (77 U/mL). A response surface methodology was used for high α-amylase production (880 U/g) by *Trichoderma virens* under optimized conditions using watermelon rinds waste biomass [44]. These findings suggest that agro-industrial crop and food wastes can be used as inexpensive raw materials for industrial amylase production, replacing the cost-intensive synthetic media.

3.2. Proteases

Proteases are amongst the most significant commercial enzymes with wide applications in food, dairy, detergent, pharmaceutical, and leather industries. Some bacterial strains of the genus *Bacillus* and many fungal strains, including *Rhizopus*, *Aspergillus*, *Penicillium*, etc. have been reported to be the most active protease producers [46,47]. The alkaline protease enzyme covers about 65% of the global enzyme market that can proficiently convert proteins to biopeptides [48]. A thermophilic fungi, *Mycothermus thermophilus*, from hydrothermal springs was used for protease production under SSF using wheat bran with an improved enzyme production (1187.03 U/mL) under optimal conditions [49]. Similarly, microbial protease was produced from a newly isolated *Neurospora crassa* under SSF which used soybean okara waste as the substrate under optimal fermentation conditions [50]. A high protease activity of 1959.82 U/g was achieved with optimal activity at pH 9 and 55 °C and preferably hydrolyzed casein protein. Camargo et al. [51] used processed orange and grape wastes for determining their bromatological characteristics and production of proteases from them employing *Aspergillus niger* CBMAI 2084. The mixed grape wastes showed specific protease activity of 174.94 U/mg, whereas a protease activity of 16 U/g·min^{-1} was observed in fermented orange waste.

Additionally, the microbial proteases produced from agro-food wastes can be exploited for various biorefining applications in addition to their potential usage in numerous food applications. Rawoof et al. [52] evaluated the effect of *Lactobacillus manihotivorans* lactic acid production from food waste by simultaneous saccharification and fermentation with high substrate utilization and less processing time. The *Lactobacillus* sp. produced appreciable protease and α-glucosidase enzymes during the hydrolysis of complex molecules in the food waste. The valorization of food waste biomass and in situ enzyme production in such a biorefining strategy could significantly decrease the operating costs of the bioprocess as compared to current industrial practices that use expensive substrates.

3.3. Lipases

Lipases, also called triacylglycerol hydrolases (EC 3.1.1.3), are vital enzymes used in numerous industrial food processes [53,54]. These enzymes are involved in various biochemical reactions that improve product quality, durability, and solubility, and provide superior organoleptic features [55]. Lipases primarily hydrolyze triglycerides to obtain free fatty acids, glycerol, monoacylglycerols, and diacylglycerols [56]. Alternatively, they catalyze the synthesis of new products via aminolysis, alcoholysis, acidolysis, esterification, and transesterification methods [17]. Lipases can be simply produced from lignocellulosic feedstocks and waste from other sources by using various microorganisms. The majority of the previous research studies have concentrated on the production of extracellular lipase with high activity by a range of microbial strains, including fungi, yeast,

and bacteria, employing both SSF and submerged fermentation (SmF) [57]. Putri et al. [57] performed lipase production from *Aspergillus niger* under optimized conditions by SSF of rice bran and *Jatropha* seed cake. The results exhibited the yield of a very dry lipase extract (282 U/mL enzyme) using 1% NaCl and 0.5% Tween 80 as the best extractants. Similarly, Pereira et al. [56], using industrially processed mango peel and seed waste, evaluated the lipase production using *Yarrowia lipolytica* by SmF. A lipase production as high as 3500 U/L of extracellular lipase was achieved under optimum conditions of temperature (27.9 °C), pH (5.0), and substrate concentrations.

Lipases can be easily produced using microorganisms under SSF by employing lignocellulosic residues as cheap feedstocks. A recent study investigated the production of lipase through *Penicillium roqueforti* growth via SSF using cocoa bran residues [53]. A maximum lipase activity of 33.33 ± 3.33 U/g was achieved using palm oil (30%) after 72 h of fermentation with the fungus. Likewise, in a promising experiment, *P. roqueforti* was used to optimize lipase production (48 U/g) employing inexpensive cocoa shell waste biomass applying an artificial neural system combined with a genetic algorithm system [54].

3.4. Lignocellulolytic Enzymes

Lignocellulolytic enzymes such as cellulases, xylanases, lignin peroxidases, laccases, etc. have received much attention in the past decade for use in the lignocellulosic biorefineries for the production of biofuels and other green organic solvents [13,14,36,58]. These enzymes have the capacity to break the complex linkages between polysaccharides (cellulose, hemicellulose) and lignin and convert them into simpler forms [35,39,59]. Due to their ability to assist in the disruption of the lignocellulose structure by degrading cellulose's β-1,4 linkages, these enzymes can improve the nutritional value of feed items with a high fiber content.

3.4.1. Cellulases and Xylanases

Cellulases are the most important class of lignocellulolytic enzymes comprising exoglucanase (E.C. 3.2.1.176), endoglucanase (E.C. 3.2.1.4), and β-glucosidases (E.C. 3.2.1.21) [24,25]. These enzymes are essential for the hydrolysis of plant biomass as they cause complete cellulose hydrolysis by their consecutive actions to form a glucose monomer for bioethanol production [60]. Additionally, cellulases have been widely applied in brewing, bread, detergents, textiles, pulp, and paper industries. Numerous bacterial and fungal species have been reported to produce cellulases using cellulose rich agro-industrial food wastes. Srivastava et al. [61], reported improved cellulase production by a novel fungus, *Cladosporium cladosporioides* NS2, under SSF employing sugarcane bagasse. The fungal isolate exhibited enhanced cellulase production with maximum filter paper cellulase (16.9 IU/gds), endoglucanase (150 IU/gds), and β-glucosidase (200 IU/gds) activity. In another study, Leite et al. [62] carried out the simultaneous production of cellulase and xylanases from different fungal strains using brewer's spent grain as cheap agro-industrial waste. *Aspergillus niger* was the best producer of the β-glucosidase enzyme with 94 ± 4 U/g activity, while *A. ibericus* achieved maximum cellulase and xylanase activities of 51–62 U cellulase/g and 300 to 313 U xylanase/g, respectively.

In the last few years, the research on xylanases has risen to the forefront due to their wide range of potential applications in numerous industries, including dairy, food, bakery, feed, paper and pulp, and lignocellulosic biorefineries [24,36]. The microbial xylanases are favored among the different sources of xylanolytic enzymes because they may possess desirable processing features and may be produced in large quantities efficiently and economically. Recently, Intasit and co-workers [63] produced a fungal xylanase from lignocellulosic palm wastes by combining SSF and SmF using *Aspergillus tubingensis* TSIP9 in a bioreactor. The combinatorial SSF–SmF endorsed higher xylanase production with high purification (7.4-fold), activity, and stability at different pH (3–8) and temperatures (30–60 °C). Singh et al. [64] conducted a study to explore rice straw, wheat straw, sugarcane bagasse, sawdust, cotton stalk, and rice husk waste as carbon sources for improved xylanase

and cellulase production by *Aspergillus flavus* under SSF. Rice straw was determined to be the best waste biomass for use as a carbon source for the production of enzymes with maximum xylanase 180 IU/gds, CMCase 235 IU/gds, FPase (12.5 IU/gds) and β-glucosidase 190 IU/gds, activities under optimum conditions. These studies demonstrated that strain microbial strains can potentially be used for cellulase and xylanase production that provides an economical method to produce high-value enzymes using agro-food wastes by SSF and SmF techniques.

3.4.2. Lignin Degrading Enzymes

Lignin is an abundantly present complex aromatic heteropolymer, which is one of the three major components of lignocellulosic biomass [16]. The lignification process is achieved through polymer–polymer coupling reactions by free radicals formed by oxidases or by the cross-linking of lignin monomers [65]. However, lignin degradation and depolymerization is necessary in many aspects for effective biomass valorization into bioproducts and carbon recycling. The agro-food waste is majorly represented as lignocellulosic biomass that acts as an ideal substrate for the action of microbes and the production of lignolytic enzymes in the food industry. Lignin peroxidases, laccases, and manganese peroxidases have been considered as suitable biological catalysts for lignin depolymerization and degradation into lignin monomers.

In a recent study, two thermotolerant lignin-degrading *Bacillus* sp. LD2 and *Aneurinibacillus* sp. LD3 exhibited an improved lignin degradation rate (61.28%) with high lignolytic enzyme activities. The lignin degrading enzymes, such as laccase, manganese peroxidase, and lignin peroxidase from these bacteria possessed maximum activities of 1484.5, 1770.75, 3117.25, and U L^{-1}, respectively [66]. Bagewadi et al. [67] used rice straw, corn cobs, sugarcane bagasse, wheat bran, and groundnut shells as potential substrates for laccase production under SSF using *Trichoderma harzianum*. The results revealed wheat bran to be suitable feedstock for maximum laccase production (510 U/g) with an 8.09-fold increase under optimized conditions. It is pertinent to note here that the high-cost commercial enzyme production systems have limited their wide scale applications in the lignocellulosic biorefineries. Therefore, the development of indigenous enzyme production processes using agro-food wastes as the inexpensive raw materials may help in bringing down the overall costs of production of second-generation biofuels and platform biochemicals.

3.5. Pectinolytic Enzymes

The pectinolytic enzymes have been widely explored in the food industry that catalyze the fragmentation of pectin-containing biomaterials forming an integral component of plant cell walls [68]. Pectin disintegration requires a set of pectinolytic enzymes including exo- and endo-polygalacturonases, and pectate- and pectin lyases [17]. Pectinases find applications in wine and fruit juice making to clarify the final product and eliminate turbidity. By using de-esterification and depolymerization reactions, the pectinases break down complex pectins in a sequential and synergistic fashion [69,70]. Most of the microorganisms can produce pectinase enzymes, however, fungi are favored for commercial applications since more than 90% of the enzyme produced is secreted into the culture medium by fungi.

Among different fungal species, *Penicillium*, *Aspergillus niger*, *Trichoderma viride*, and *Rhizopus* offer many advantages as pectinase producers since they are designated as Generally Regarded as Safe (GRAS) strains and produce extracellular bioproducts that may be readily recovered from the fermented medium [68]. Núñez Pérez et al. [69] isolated a wild strain of *Aspergillus* sp. To produce pectinases from pectin-rich dehydrated coffee pulp and husk under SSF with 29.9 IU/g of enzyme activity. Sethi et al. [70] reported the use of natural substrates, such as neem oil cake, mustard oil cake, groundnut oil cake, green gram peels, black gram peels, chickling vetch peels, pearl millet residues, broken rice, wheat bran, finger millet waste, apple pomace, banana peels, and orange peels for pectinase biosynthesis from *Aspergillus terreus* NCFT4269. The results displayed that a maximum pectinase activity of 6500 ± 1116.21 U/g was achieved under SSF, while the

liquid static surface fermentation gave the enzymatic activity of 400 ± 21.45 U/mL under optimized conditions. Future research studies on pectinolytic enzymes should be focused on determining the molecular processes that control enzyme secretion in addition to modes of action of diverse pectinolytic enzymes on agro-food pectic substrates. It would offer a strong platform for controlling microorganisms to produce large amounts of effective and affordable enzyme systems.

4. Low-Cost Enzyme Immobilization Strategies

The bioconversion of agro-industrial food and crop waste into valuable fuels and bioproducts often depends on the cost of the method, equipment, and infrastructure, as well as the market value of the finished products. Enzymes offer an array of advantages in the valorization of abundantly generated agro-food wastes [19,21,41,42,69]. They perform biochemical catalytic reactions with exceptional specificity and decent stereo-selectivity, and, under very mild reaction conditions, enable the development of more environmentally friendly, green, and sustainable biochemical processes [21]. The operating range of enzymes is, however, rather constrained, and enzymes originating from natural sources are particularly effective only under their optimal conditions [18]. Moreover, a lack of maintenance of their stability, catalytic activity, and recovery under variable industrial bioprocessing conditions of pH, temperature, water activity, and solvent properties is a major hurdle [71]. Therefore, the development of steady biocatalytic systems involving enzymes is crucial for expanding their commercial applications. In the last few years, enzyme immobilization has received significant attention as an important bio-engineering approach to customize and enhance a wide range of enzyme catalytic characteristics, including activity, physicochemical stability, specificity, selectivity, and tolerance of inhibitors [12,72,73]. The overall cost of the enzymatic production process can be reduced by immobilizing the industrially significant enzyme support matrixes [74]. To create reusable, long-standing, and stable immobilized biocatalytic systems, the choice of the supporting matrix and the immobilization technique is essential [4]. A number of emerging support materials have been recently used for enzyme immobilization, such as magnetic nanoparticles, graphene oxide, polyurethane foam, or chitosan [75]. The inexpensive immobilization matrixes possess several advantageous properties that help in developing highly efficient biocatalysts (Figure 4).

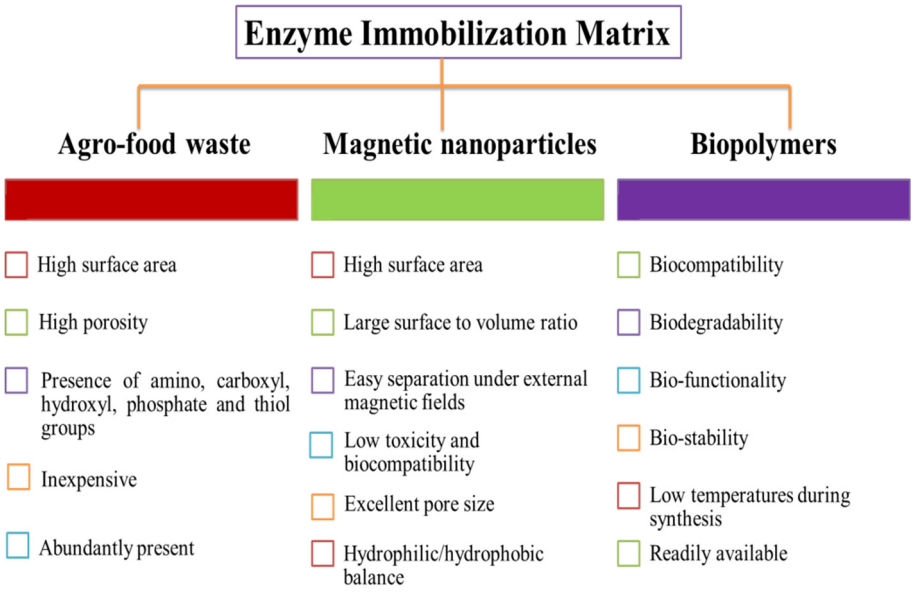

Figure 4. Properties of different enzyme immobilization matrixes.

These support materials can be used to immobilize and entrap the enzymes through different techniques, such as adsorption, crosslinking, covalent binding, and

entrapment [21,75–79]. Table 1 shows the list of promising enzyme immobilization supports with major results and applications.

Table 1. Enzyme immobilization by different supporting materials.

Supporting Material	Immobilization Method	Immobilized Enzyme	Major Results	Application	Reference
Rice husk ash	Adsorption	Lipase	Higher adsorption capacity of biocatalyst	Biodiesel production	[80]
Magnetic rice straw (MRS)	Adsorption	Lipase	36% higher esterification yield with Lipase-MRS composite	Biodiesel production	[73]
Rice husk	Covalent	Lipase	Immobilized lipase exhibited high esterification yield (88.0%)	Esterification reaction for biodiesel production	[81]
Rice husk, sugarcane bagasse, babassu mesocarpus, corn cobs, coffee grounds, coconut bark	Adsorption	Lipase	High immobilization efficiencies (>98%)	Hexyl laurate production	[77]
Green coconut fiber	Adsorption and cross-linking	Laccase	>80% initial activity retained up to six cycles	Oxidation of aromaticorganic and inorganic compounds	[82]
Spent coffee grounds	Covalent	β-glucosidase	Enriched aglycone content (67.14 ± 0.60%) by enzymatic treatment	Isoflavone conversion in black soy milk	[83]
Spent grain	Adsorption	Trypsin	High operational and thermal stability of immobilized enzyme	Protein hydrolysis	[84]
Rice straw biochar	Adsorption	Laccase	Increased enzyme stability and reusability	Anthracene biodegradation.	[85]
Rice husk, egg shell membrane	Covalent	Lipase	High immobilization efficiency for rice husk (81%) and eggshell membrane (87%)	-	[86]
Tomato peels	Covalent	Pectinase	High thermal and pH stability, reusability, and storage stability of immobilized biocatalyst	Lycopene production	[87]
Fruits peels and scraps	Covalent	Pectinase and cellulase	80% of residual activity retained for magnetic biocatalyst after ten cycles	Antioxidants production	[88]
Dairy waste	Cross-linkage	Lactase, glucose isomerase	Improved transglycosylation activity of enzyme for improve lactulose yield	Prebiotics production	[89]
Green coconut fiber	Covalent	Laccase	100% activity after 10 cycles	Clarification of apple juice	[90]
Chicken Feather	Covalent	Laccase	94.32% after 3 weeks of storage	Oxidation of Veratryl alcohol	[76]
Carboxymethyl cellulose nanoparticles	Covalent	Lipase	Immobilized enzyme with higher activity and stability	Diverse applications in food industry	[91]
Dialdehyde starch nanoparticles	Adsorption	Lipase	High stability (82.5%) and recycling rate (53.6%)	Food processing	[92]
Graphene oxide-magnetite naoparticles	Covalent	α-amylase	Enhanced stability and half life of immobilized enzyme	Maltose containing syrup production	[93]
Alginate beads	Covalent	Pectinase	Improved thermostability of immobilzed enzyme	Juice processing	[94]
Alginate-gelatin hydrogel matrix	Cross-linkage	Lipase	High thermal stability (96% activity)	Fatty acid production	[79]

4.1. Agro-Food Wastes for Enzyme Immobilization

The exploitation of agricultural residues and food wastes as novel immobilization or supporting matrices for enzymes offers economic solutions for industrial bioprocess, in addition to solving the issues of environment pollution and waste disposal. Undeniably, agro-industrial food and crop wastes contain a variety of characteristics with intriguing potential applications, including high surface area, high porosity, and the presence of several chemical groups, such as amino, carboxyl, hydroxyl, phosphate, and thiol groups [95]. Otari et al. [73] reported a one-step novel and robust method of lipase immobilization on magnetic rice straw using Fe_2O_3 nanoparticles. The results exhibited high lipase immobilization efficiency of 94.3% with 91.3 $mg \cdot g^{-1}$ of enzyme loading, increased enzymatic stability by 8-fold at a high temperature (70 °C), and reusability. Lira et al. [77] used rice husk, sugarcane bagasse, babassu mesocarp, corn cobs, coffee grounds, and coconut bark residual biomasses as supports for lipase immobilization extracted from *Thermomyces lanuginosus*. The results showed an immobilization efficiency of more than 98% with high hydrolytic activity of 4.608 U/g using rice husks as immobilization supports. The activation of agro-industrial food wastes as supports/carriers for enzyme immobilization with low-cost materials postures an upper hand for researchers with its constant availability and environment friendliness. Recently, the stability of *Candida rugosa* lipase was enhanced by its covalent immobilization by glutaraldehyde-activated agricultural wastes. Rice husk support provided the highest stability to the enzyme with the highest retention of initial activity (94.1%), followed by sugarcane bagasse (90.3%) and coconut fiber carrier (89.3%) [81].

Lignocellulolytic enzymes have also been immobilized using agro-food based biomasses as support materials. In a previous study [85], the ligninolytic enzyme-laccase was immobilized on the surface of rice straw biochar for anthracene biodegradation. The laccase exhibited an effective immobilization yield of 66% with high stability up to six cycles and retention of 40% of initial activity. Similarly, Ghosh and Ghosh [82] successfully immobilized purified laccase from *Aspergillus flavus* PUF5 on coconut fiber while retaining 80% of its initial activity after using it for six repeated cycles. These findings support the notion that agro-industrial crop and food waste biomass can produce stable and robust biocatalysts containing immobilized industrially important enzymes with better results than commercial preparations.

4.2. Magnetic Nanoparticles for Enzyme Immobilization

The development of strong enzymatic systems with high stability and catalysis at several extremities is required for commercial applications. Magnetic nanoparticles are regarded as potential support materials when compared to conventional immobilization carriers because of their high surface area, small size, and large surface to volume ratio [74,75]. The nanostructural support materials have huge potential to develop nanobiocatalyst systems involving enzymes displaying high catalytic properties in both aqueous and non-aqueous environments [96]. Suo et al. [91] immobilized lipase on ionic liquid-modified magnetic carboxymethyl cellulose nanoparticles that exhibited strong specific activity, which was 1.43 fold higher than that of the free lipase enzyme. In similar research, Yang et al. [92] achieved high storage stability (82.5%) and recycling rate (53.6%), and better stability and durability of lipase enzymes immobilized by magnetic dialdehyde starch nanoparticles. Moreover, the immobilized lipase displayed better enzymatic properties and improved acid-base tolerance and thermal stability as compared to the free enzyme.

In another research study, Desai et al. [93] prepared graphene oxide–magnetite nanoparticles for amylase immobilization through covalent bonding, which increased the half-life of the immobilized enzyme (20 h) as compared to the free enzyme (13 h). The immobilized amylase also demonstrated high reusability up to eleven subsequent cycles during the production of high maltose containing syrup. The co-immobilization strategy by nanoparticle composites involving two or more enzymes have also been attempted. Nadar and Rathod [88] simultaneously co-immobilized pectinase and cellulase enzymes onto amino functionalized magnetic nanoparticles for antioxidants extraction from waste fruit peel

residues. When compared to free form, the magnetic nano-biocatalyst demonstrated a two-fold improved half-life in the temperature range of 50–70 °C and retained up to 80% of residual enzyme activity even after ten repeated cycles. Immobilized laccase on Fe_3O_4 magnetic nanoparticles was employed in a novel detoxifying method to enhance *Rhodotorula glutinis* lipid synthesis from rice straw hydrolysate [96]. The immobilized laccase presented better stability, retaining 56% of its original activity at pH 2 and 76% at 70 °C compared with the free laccase. These findings suggest that enzyme immobilization on magnetic nanoparticles has immense potential to valorize the agro-food waste hydrolysate for improving the production of valuable bioproducts.

4.3. Biopolymers for Enzyme Immobilization

Biopolymers are quite valuable among supports for enzyme immobilization because of their 'green' properties, such as biocompatibility, biodegradability, bio-functionality, and bio-stability [74]. Additionally, biopolymers offer a wide range of chemical and structural properties, need low temperatures during synthesis, and are readily available [97]. Chitin and chitosan are remarkable immobilization carriers of interest among the support materials explored for immobilizing enzymes. A study was conducted on immobilization of pectinase onto chitosan magnetic nanoparticles using dextran polyaldehyde as a macromolecular cross-linking agent [78]. The immobilized pectinase showed high reusability and permanence retaining 85% and 89% of its original activity after seven cycles and fifteen days of storage, respectively. Recent reports suggest the use of new immobilized food enzymes in food applications. In a novel and sustainable method, a fluidized bed reactor with immobilized propyl endopeptidase protease enzyme from *Aspergillus niger* on food-grade chitosan beads was used to continuously generate high-quality, gluten-free beer from barley malt [98].

The use of hydrogel matrices, such as alginate, for enzyme immobilization is quite popular because they are environmentally benign enzyme carriers with excellent gel porosity and biocompatibility. Recently, Abdel-Mageed et al. [79] synthesized an alginate-gelatin hydrogel matrix as immobilization support for lipase from *Mucor racemosus* (Lip). The immobilized biocatalyst exhibited improved thermal stability while retaining 96% of its activity after four cycles and 90% of its original activity at 4 °C stored for 60 days. Likewise, *Aspergillus aculeatus* pectinase immobilized on alginate beads ensured satisfactory thermostability of the enzyme with low values of activation entropy for juice processing [94]. A recent study focused on the use of co-immobilized pectinase, amylase, and cellulase using different immobilization matrices, such as chitosan, silica gel, and sodium-alginate for fruit juice clarification. The highest activity for amylase (90.9 $\pm$ 4.3 U/g carrier), pectinase (85.7 $\pm$ 4.0 U/g carrier), and cellulase (23.2 $\pm$ 1.1 U/g carrier) was observed when they were co-immobilized onto silica gel with immobilization efficiency of 67.9%, 53.6%, and 72.9%, respectively [99]. These research developments suggest the potential of these biopolymers in enzyme immobilization processes in paving the way for wide scale applications of enzymes in food analysis, food bioprocessing, and food control, among other areas.

5. Enzyme Engineering Approaches

Though enzymes catalyze a wide variety of biochemical reactions, yet they are not suited for many essential catalytic bioprocesses or other industrially relevant substrates that are beyond their natural cellular micro-environments. The desired attributes of diverse industries can be satisfied by using tailored enzymes in novel, cutting-edge enzyme engineering and stabilization techniques, opening up new opportunities for their use in biocatalysis. In the present scenario, the high value of protein engineering has been well recognized in industrial-level biotransformation [100]. Protein engineering, with assistance from molecular approaches or directed evolution, rational design, or computational methods, enables the accelerated designing of biocatalysts that are ideal for any desired

bioprocess with commercial applications [101]. Figure 5 explains the different strategies of enzyme engineering for improved biocatalysts.

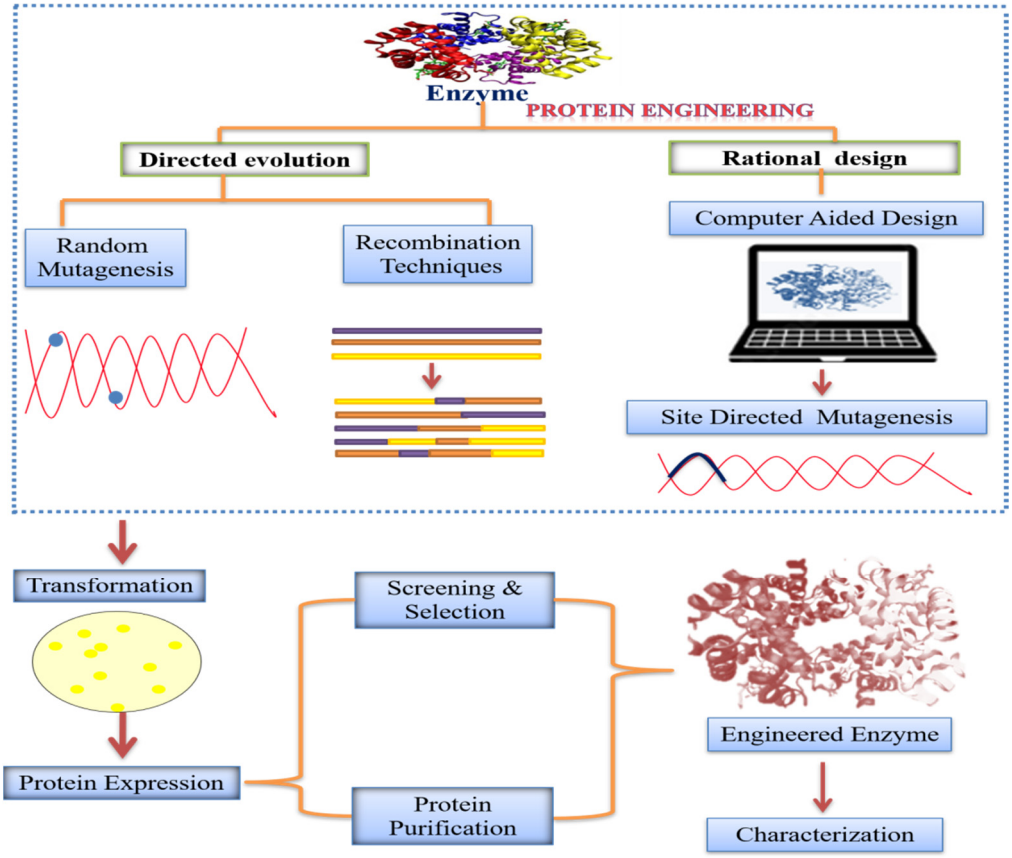

Figure 5. The schematics of different enzyme engineering methods.

The enzymatic diversity created by these methods through the application of semi-rational designs based on artificial intelligence, protein structure, and sequence information yields superior specificity and functionality of industrial enzymes. Different approaches for the enzyme engineering are presented in Table 2.

Table 2. Different enzyme engineering strategies and engineered biocatalysts.

Method	Enzyme	Source	Improved Properties	References
Directed evolution	α-amylase	*Bacillus cereus* GL96	Higher pH and thermostability	[102]
	β-glucosidase A	*Clostridium thermocellum*	Higher thermostability and catalytic activity	[103]
	Lipase	*Proteus mirabilis*	4.3-fold catalytic efficiency and enhanced stability of enzyme	[104]
	Amylase	*Bacillus licheniformis* R-53	Enzyme with better effects in delaying recrystallization, reducing hardness, improving elasticity of bread dough	[105]
	β-glucosidase	*Pichia pastoris*	Better saccharification efficiency	[106]

Table 2. *Cont.*

Method	Enzyme	Source	Improved Properties	References
Rational design	Serine peptidase	*Pseudomonas aeruginosa*	High thermostability and catalytic activity	[107]
	Lipase	*Candida rugosa*	Higher enzyme esterification yield (88.0%) and retained 72.7% of initial activity	[81]
	β-glucosidase	*Talaromyces leycettanus*	Improved substrate affinity and catalytic efficiency	[108]
	α-amylase	*Bacillus licheniformis*	High specific activity and thermostability (70 °C)	[109]
	α-amylase	*Bacillus subtilis*	Improved hydrolytic pattern of engineered enzyme	[110]
Computational designs	Lipase	*Rhizopus chinensis*	High catalytic efficiency (700%), potential for propeptide to shift the substrate specificity	[111]
	Lipase	*Yarrowia lipolytica*	Increased reaction rate and enzyme recyclability	[112]
	Lipase	*Pseudomonas aeruginosa*	Increased enzyme stability and activity by creation of α-helix hotspots after immobilization	[113]
	Glucose oxidase	*Aspergillus niger*	Improved catalytic efficiency with high gluconic acid yield (324 g/L)	[114]

5.1. Directed Evolution

The enzyme biocatalysts must exhibit superior productivity and selectivity with minimal catalyst loadings at high substrate concentrations, which can be accomplished by creating mutant enzyme libraries and using directed evolution. Directed evolution (DE) is a potent method for producing efficient enzyme catalysts that can carry out a variety of biocatalytic tasks without in-depth knowledge of structure–function correlations [115]. DE, also referred to as molecular evolution, does not require information of the sequence or three-dimensional structure of the enzymes [116]. Consistent high-throughput screening of the mutant library and effective library construction are required for successful DE experiments. The major highlights of DE include the creation of random gene libraries, expression of genes in an appropriate host, and screening libraries of mutant enzymes according to the need [117].

Recently, Pouyan et al. [102] used an in silico approach to redesign α-amylase from *Bacillus cereus* GL96 for higher thermostability and characterized the engineered enzyme using directed evolution. The engineered α-amylase exhibited superior properties over wide temperature (70 °C) and pH (4–11) ranges. In another study, lipase from *Proteus mirabilis* was genetically fused to self-crystallizing protein (Cry3Aa) for immobilized lipase crystal production by DE in *Bacillus thuringiensis* [104]. The immobilized lipase mutant exhibited 4.3-fold greater catalytic efficiency and enhanced stability that could efficiently catalyse waste cooking oil into biodiesel for at least 15 cycles with reasonable conversion efficiency.

Similarly, in another study, the site-directed mutagenesis of a lipase from *Pseudomonas fluorescens* provided enhanced thermostability to the enzyme which could be applied to food applications [118]. The transglutaminase enzyme from *Streptomyces mobaraensis*, which catalyses the cross-linking modification of proteins and other biotechnological fields, was subjected to site-directed mutagenesis [119]. The variant enzyme possessed higher thermostability with improved specific activity. These studies provide deeper

insights of the structure–function relationship for improving the thermostability of different enzymes through directed evolution. It also offers a theoretical framework and background knowledge for designing enzymes with improved properties to satisfy industrial demands.

5.2. Rational Design

Rational design (RD) is a classical protein engineering strategy for obtaining tailored enzymes with improved catalytic properties, kinetics, thermostability, substrate specificity, and resistance to organic solvents [107,116]. RD brings precise variations in the amino acid sequence via site-directed mutagenesis and is used when the structure, function, and mechanism of action of the target enzyme is already known [120]. The enzyme of interest can be engineered by RD involving targeted mutagenesis, computational techniques, and a de novo design [121]. In a study by Ashraf et al. [107], a serine peptidase from *Pseudomonas aeruginosa* was engineered using rational design for improved thermal stability and catalytic efficiency. The mutant enzyme exhibited higher T_m and increased residual activity at elevated temperature compared to the wild type.

Some researchers have recently used computational design techniques to alter important industrial enzymes used with improved enzymatic activity. Costa et al. [122] employed a computational protein design method to redesign Cel9A-68 cellulase from *Thermobifida fusca* through linker mutations that facilitated higher enzymatic activity for cellulose degradation. In another study by Elatico et al. [113], the computational technique was used to reverse engineer the lipase from *Pseudomonas aeruginosa* PAO1 using proline mutations. The technique helped in creating variants with possible α-helix hotspots for augmented enzyme activity and stability. Given the promising traits the mutant enzyme displays, these protein engineering methods could be taken into consideration for additional research to fulfil the industrial requirements.

6. Current Challenges and Future Prospects

Sustainable development based on the idea of a circular economy could possibly assist in achieving the targets of global waste minimization, valorization, and its recycling. The agro-industrial food waste which is an underutilized resource ideally fulfills the criteria for circular economy for conversion into useful bioproducts. Agro-food waste contains a significant amount of latent nutrients that can be efficiently extracted, recycled, repurposed, and used as substrates for enzyme production. Enzymes have been widely explored in the food industry and lignocellulosic biorefineries for producing numerous value-added biochemicals. However, the scaling up of enzyme production still faces a huge research gap to meet the industrial requirements. The significant challenges and barriers, including high production costs, low stability, and long reaction times, among others, still persist in the commercial applicability of enzymes. Moreover, the market cost of enzymes is quite high owing to the fact that expensive synthetic substrates and processes are used for their production. For enzyme prices to be competitive, they would need to be an average of $0.10 per gallon [123]. The development of biocatalytic enzyme systems from low-cost agro-food wastes represent a distinctive technological approach for environmental and economical sustainability. Different strategies to improve enzyme production costs have been proposed through comprehensive research efforts over the past few decades. Additionally, the shortlisting of agro-food wastes as the carriers for enzyme immobilization with various operational requirements is challenging, but exciting in terms of further mitigating the cost-related issues of enzyme applicability at industrial levels.

There are certain challenges also related to enzyme immobilization practices. These include enzyme distortion during immobilization, steric hindrance of enzymes with substrate, rapid consumption of the substrate, etc. The distortion of the enzyme during immobilization occurs when the enzyme is being used under more severe conditions than normal conditions. However, stabilizing the enzyme during immobilization might allow for higher activity than the soluble enzyme. The steric interference of enzymes with the substrate may depend upon the enzyme loading on the immobilization support. If the surface of the

support material does not completely block the active site, a reasonable activity against large substrates can be found with minimal enzyme load, resulting in enzyme molecules with free space around them to bind with the substrate. Apparently, the substrates with different molecular sizes and using different enzyme loadings could help in understanding if the issues are caused by steric hindrances or enzyme distortion. In certain cases, the enzyme is physically adsorbed on the surface of the immobilization matrix and may release from the support, resulting in lower efficiency. This issue may be discovered by measuring the activity in the washing solutions, particularly the initial ones. Therefore, a deeper understanding of the mechanisms of enzyme immobilization on the support matrixes is necessary to mitigate these inadequacies.

Looking forward, the improvements in the science and engineering knowledge for choice of microorganism, enzyme production under SSF and SmF systems, and maintenance of optimum chemical, physical, and biological parameters could develop the sustainable bioprocess. The development of stable biocatalytic systems by novel immobilization technologies using agro-food wastes as carriers could also elevate the industrial applications of enzymes. Over the last several years, the increase in the market demand for enzymes to establish new technological bioprocesses has substantially driven the need for engineered enzymes with unique biocatalytic and economic attributes. Research on engineering of the local enzyme environments and their catalytic regions using exciting computational and machine learning technologies is expected to further increase in coming years and involve multi-step reaction cascades, economizing the overall bioprocess. Moreover, powerful tools like life-cycle assessments and techno-economic analyses could be used for the evaluation of the viable commercial-scale biocatalytic processes. Further understanding of enzymes can be more effectively used in a range of industrial processes, which will come from research studies of both known and yet-to-be discovered enzymes.

7. Conclusions

The management of food and agricultural trash is one of the most pressing issues for modern civilization. The proper repurposing of agro-food wastes utilizing green technology is critical to reduce the negative and destructive consequences of waste disposal that produce compounds with added value, aiding towards implementing circular economy. Microbial enzymes play a key role in the valorization of agro-industrial crop and food wastes compared to conventional chemical catalysts. The utilization of agro-food waste to produce commercially important enzymes by microorganisms offers great promise for efficient waste utilization and sufficient biocatalytic systems with high conversion efficiencies, thereby allowing achievement of the targets of sustainable development. Furthermore, using novel, inexpensive enzyme immobilization supports and engineered enzymes can exhibit improved catalytic performance when applying them to industrial food applications.

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Abbreviations

AW	Agricultural waste
FAO	Food and agriculture organization
FVW	Fruit and vegetable waste
GRAS	Generally Regarded as Safe
KW	Kitchen waste
LCB	Lignocellulosic biomass
SmF	Submerged fermentation
SSF	Solid state fermentation

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