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Review

Ionizing Radiation Technologies to Increase the Extraction of Bioactive Compounds from Agro-Industrial Residues: A Review

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ABSTRACT: Due to the growing demand in society for healthier foods, scientific communities are searching and developing new ingredients. In this context, agro-industrial residues, which can have a negative impact on the environment, represent a natural source for bioactive compounds and their recovery can contribute to economic and environmental sustainability. Ionizing radiation is a clean and eco-friendly technology that can be used to improve the extraction of bioactive compounds. The aim of this review, after presenting general aspects about bioactive compounds in agro-industrial residues and radiation technologies, is to focus on the effects of ionizing radiation on the extraction of bioactive compounds from these residues and related bioactive properties. Irradiated residues were demonstrated to have enhanced bioactive characteristics that turn the prepared extracts suitable for applications in food industry, resulting in high-added-value products as well as reducing adverse impacts on the environment.

KEYWORDS: *agro-industrial residues, extraction, bioactive compounds, bioactivities, ionizing radiation*

1. INTRODUCTION

The agro-industrial residues can be many and different wastes from agriculture and industry. Agriculture residues can be divided into field residues and process residues and can include leaves, stalks, seedpods, stems, molasses, husks, pulp, and peels.[1] These wastes can also comprise the byproducts of the agro-food industry such as oil cakes from oil extraction, degummed fruits and legumes, coffee dregs, milk serum, sludge from wool, cellulose, bran, starch, juice, and sugars.[2]

With the fast developing world, the high volume of wastes produced by these industries generates large quantities for disposal and represents a loss of underutilized biomass and nutrients. Although some of them can be discharged safely, others are considered to have harmful effects on the environment with consequences to human and animal health. This problem imposes the need to find strategies to reuse the valuable agro-industrial residues in a more efficient way, providing environmental benefits and contributing to the economic sustainability. The agro-industrial residues can be used as fertilizers in agriculture, biofuel and enzyme production, preparation of biodegradable polymeric systems, recycled agricultural composting and extraction of food flavoring, preservative and bioactive compounds.[3]

Nowadays, consumers are more conscious and interested in what they eat, thus increasing the demand for healthier foodstuffs.[4] In this way, the food industry tried to answer this challenge and started to develop innovative and natural functional products[5] that could be used to replace the existing synthetic ones or as new added-value ingredients. The extractable antioxidant and antimicrobial compounds from agro-industrial residues offer a more natural and safer alternative to the food industry.[6]

The purpose of this review is to provide an overview of the bioactive compounds recovery from agro-industrial wastes to develop functional/health foods. The current status of the extraction of agro-industrial bioactive compounds is summarized, and for the first time, we discuss the positive impact of the ionizing radiation technology on the extractability of these compounds.

2. EXTRACTION OF BIOACTIVE COMPOUNDS

Bioactive compounds are components naturally present in plant and food products that provide health benefits and wellness, including antioxidant, anti-inflammatory, antimicrobial, anticancer, and immunomodulatory activities.[7] Natural bioactive compounds have diverse structures and functionalities with molecules having enormous potential for the production of nutraceuticals, functional foods, and food additives.[8] With this purpose, these compounds are being intensively studied to evaluate their effects on health, showing benefic effects for cardiovascular disease, cancers, and others. They comprise compounds with different chemical structures (hydrophilic or lipophilic), distribution in nature, range of concentrations, possible site of action, effectiveness against oxidative species, specificity, and biological action. The main classes of bioactive compounds include polyphenolic compounds, carotenoids, tocopherols, phytosterols, organosulfur

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compounds, fatty acids, betalains, essential oils (terpenes), and alkaloids.[7]

The extraction of these compounds can be performed by using conventional and nonconventional methods. Conventional techniques are based on solid−liquid extraction requiring the use of organic solvents, temperature, and agitation and include percolation, maceration (ME), decoction, and hydrodistillation. Some extraction variables such as type of solvent and product/solvent ratio, time and temperature of extraction, and mode of stirring are significant influencing factors on the recovery of bioactive compounds and should be considered in the process optimization.[9]

To overcome limitations of conventional methods, such as long extraction time, requirement of costly, high purity, and safe solvents, evaporation of the large amount of solvent, and/or low extraction selectivity, new and promising techniques have been introduced[10,11] such as ultrasound-assisted extraction (UAE),[12,13] pulsed electric field (PEF) extraction,[14,15] enzyme-assisted extraction (EAE),[16−18] microwave-assisted extraction (MAE),[19,20] pressurized liquid extraction (PLE),[21] supercritical fluid extraction (SFE),[22] pressurized low-polarity water extraction, and molecular distillation. M'hiri, Ioannou, Paris, Ghoul, and Boudhrioua (2016)[23] compared the efficiencies of different methods (ME, UAE, MAE, high pressure extraction (HPE), and supercritical CO_2 extraction ($SC\text{-}CO_2$)) for antioxidant extraction from *Citrus sinensis* (L.) Osbeck peel, evaluating selectivity, total phenol content, total and individual flavonoids, and antioxidant activity; they concluded that MAE was the extracting method to obtain higher contents of phenols, flavonoids, and individual flavonoids. Tabaraki and Ghadiri (2016)[24] also obtained the best results on the extraction of antioxidants from *Pistacia vera* L. hull using MAE when compared with UAE and conventional extraction. Vieira et al. (2017)[25] compared conventional maceration and MAE for the extraction of valuable compounds from *Juglans regia* L. leaves, also attributing to MAE better outcomes. On the other hand, UAE using acidified water−ethanol mixtures as solvent was considered the optimal technique to extract bioactive anthocyanin pigments from *Ficus carica* L. peel, when compared with heat-assisted extraction (HAE) and MAE.[26] In another study, the multifrequency multimode modulated (MMM) ultrasonic technique increased the recovery of phenolic compounds from olive pomace and its antioxidant activity when compared to the conventional extraction.[27]

In general, although the results regarding the technique of choice differ depending on the study, the assisted extraction methods showed to be more effective than the conventional ones to extract polyphenol antioxidants from agricultural byproducts.

3. ISOLATION AND CHARACTERIZATION OF BIOACTIVE COMPOUNDS

After extraction, the obtained extracts contain complex mixtures of natural compounds that have to be isolated, purified, and further characterized. The development of efficient methods for isolation/purification/characterization of the bioactive compounds has been an important challenge for researchers, which depends on the chemical and physical characteristics of them.[28,29]

The isolation and purification of the compounds involves different chromatographic techniques, such as thin-layer chromatography (TLC), column chromatography, flash chromatography, and high-performance liquid chromatography (HPLC). TLC is a simple, cost-effective, and rapid chromatographic technique, which gives the number of components present in the mixture but can also identify the compound comparing with a standard one, and HPLC is a versatile and widely used technique for the isolation of compounds from agro-industrial residues.[30]

The structure and biological activity of the purified compounds are then determined using different methodologies. Fourier transform infrared spectroscopy (FTIR) can be considered as a fingerprinting tool due to their unique spectra of pure compounds, allowing the identification of functional groups present in a compound.[31] Furthermore, HPLC can be coupled with simple detectors, such as UV detectors, or detectors for hyphenated systems, such as UV-diode array (DAD), mass spectrometry (MS), or nuclear magnetic resonance (NMR), producing multidimensional data for online identification and dereplication purposes.[29] An overview of the employed methodologies in extraction, isolation, and characterization of bioactive compounds from residues of agro-food industries can be found in Figure 1.

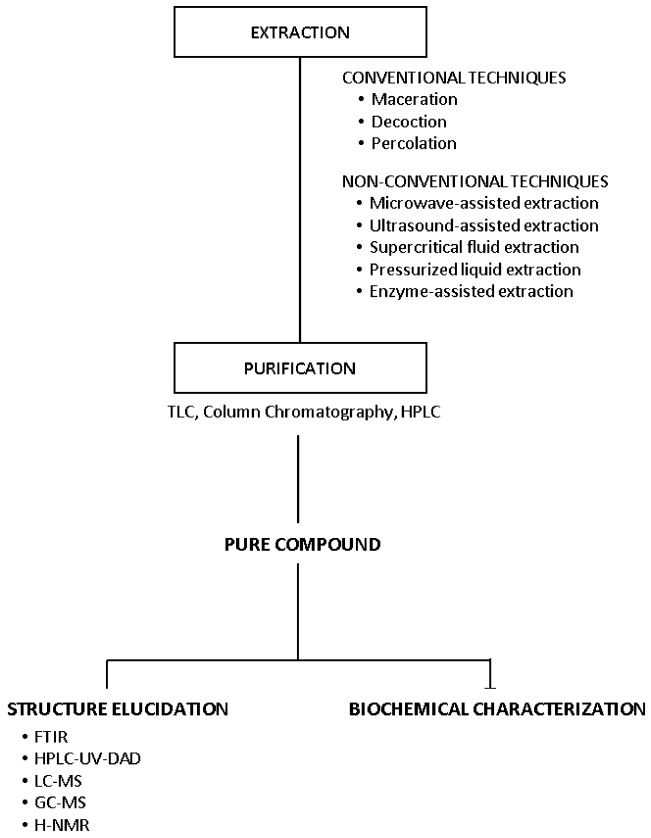

Figure 1. Methodologies used in extraction, isolation, and characterization of bioactive compounds from agro-industrial residues.

These techniques can be combined in order to provide a rapid and accurate identification of compounds, especially when the pure compound is unknown and not available. Hence, it is important to precisely select the methods to use from the extraction to the separation and purification, as this is crucial to obtain accurate results.

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Table 1. Literature Review of the Extraction of Bioactive Compounds from Agro-Industrial Residues Using Different Techniques and Conditions[a]

agro-industrial residue	extraction technique	extraction conditions: solvent and proportion		temperature (°C)	time (min)	others	main extracted bioactive compounds	references
sugar cane bagasse	ME	MeOH:H_2O	50:50	RT	>500		gallic acid; ferulic acid; epicatechin; quercetin derivatives; kaempferol derivatives	37
			70:30					
corn husk			100:0					
peanut husk		EtOH:H_2O	50:50					
coffee cherry husk			70:70					
			100:0					
rice bran		H_2O	100					
wheat bran								
Coffea arabica L.	filter coffee-maker	H_2O	100	90	6	defatted with petroleum ether	5-*O*-caffeoylquinic acid; 3,4-dicaffeoylquinic acid; 3-caffeoylquinic acid; 4-caffeoylquinic acid; 3,5-dicaffeoylquinic acid; 4,5-dicaffeoylquinic acid	99
Oryza sativa L. bran	ME	EtOH:H_2O	80:20	RT	20		protocatechuic acid; vanillic acid; feruclic acid; *p*-coumaric acid; α-tocopherol; γ-tocopherol; γ-tocotrienol; peonidin 3-glucoside; cyaniding 3-utinoside; cyaniding 3-glucoside	35
wheat bran	ME	MeOH	100	RT	>500		gallic acid; caffeic acid; *p*-coumaric acid; ferulic acid	36
		MeOH:H_2O	80:20					
		EtOH	100					
		EtOH:H_2O	80:20					
		C_3H_6O:H_2O	50:50					
barley husk	ME	MeOH	100	RT	>500		gallic acid; chlorogenic acid; caffeic acid; *p*-coumaric acid; ferulic acid; benzoic acid; quercetin; kaempferol	36
		MeOH:H_2O	80:20					
		EtOH	100					
		EtOH:H_2O	80:20					
		C_3H_6O:H_2O	50:50					
Juglans regia L. leaves	ME	EtOH	0−100	42−78	30−126		3-*O*-caffeoylquinic acid; quercetin 3-*O*-glucoside; quercetin *O*-pentoside	25
	MEA	EtOH	0−100	60−180	3−37	850 W		
Araucaria angustifolia (Bertol.) Kuntze coats	ME	EtOH	100		300		protocatechuic acid; (+)-catechin; prodelphinidin B3; B-type (epi)catechin dimer; prodelphinidin B3; (−)-epicatechin	100
		EtOH:H_2O	80:20					
	ME	EtOH	38−97	21−64	15	S/L = 2−13%		101
Eucalyptus globulus Labill. bark	ME	MeOH + H_2O	100	RT	>500		gallic acid; quinic acid; catechin; chlorogenic acid; galloyl-bis(hexahydroxydiphenoyl)-glucose; galloyl-hexahydroxydiphenoyl-glucose; digalloylglucose; isorhamnetin-rhamnoside; eriodictyol	39
		MeOH:H_2O	50:50	RT	>500			
	SFE	CO_2	100	70	225	300 bar	eriodictyol; isorhamnetim; naringenin; methyl-ellagic acid-pentose; methyl-ellagic acid	40
		CO_2:EtOH	85:15					
		CO_2:EtOAC	85:15					
		CO_2:H_2O	98:2					
Eucalyptus grandis W.Hill bark	ME	MeOH:H_2O	50:50	RT	>500		gallic acid; catechin; galloyl-*bis*-hexahydroxydiphenoyl-glucose; epicatechin; ellagic acid-rhamnoside; ellagic acid; digalloylglucose	41

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Table 1. continued

agro-industrial residue	extraction technique	extraction conditions: solvent and proportion		temperature (°C)	time (min)	others	main extracted bioactive compounds	references
Eucalyptus urograndis (*E. grandis* W.Hill × *E. urophylla S.T.Blake*) bark	ME	$MeOH:H_2O$	50:50	RT	>500		quinic acid; gallic acid; galloyl-*bis*-hexahydroxydiphenoyl-glucose; epicatechin; ellagic acid-rhamnoside; ellagic acid	41
Eucalyptus maidenii F.Muell. bark	ME	$MeOH:H_2O$	50:50	RT	>500		quinic acid; gallic acid; catechin; chlorogenic acid; methyl-ellagic acid-pentose; myricetin-rhamnoside; ellagic acid; taxifolin	41
Quercus suber L. planks	ME	$MeOH:H_2O + (C_2H_5)_2O$	80:20	RT	>500		ellagic acid; gallic acid; protocatechuic acid; caffeic acid; esculetin; salicylic acid; eriodictyol	38
		$MeOH + H_2O$	100	RT	360 + 360			
			100					
Quercus suber L. wastewater							gallic acid; protocatechuic acid; vanillic acid; syringic acid; ferulic acid; ellagic acid	42
Theobroma cacao L. pod husk	ME	$C_3H_6O:H_2O:CH_3COOH$	70:29.5:0.5	RT	60		catechin; quercetin; (−)-epicatechin; gallic acid; coumaric acid; protocatechuic acid	43
	ME	$EtOH:H_2O$	80:20	40	30		protocatechuic acid; *p*-hydroxybenzoic acid; salicyclic acid; kaempferol; linarin; luteoin; apigenin	44
Olea europaea L. leaves	PLE	EtOH	100	150	20		gluconic acid; quinic acid; luteolin-7-*O*-glucoside; luteolin-4-*O*-glucoside; oleuropein; oleuroside; lucidumoside	52
		H_2O	100	200	20		gluconic acid; hydroxytyrosol; oleoside; elonolic acid glucoside; hydroxytyrosol acetate; luteolin-7-*O*-glucoside; oleuropein; oleuroside; luteolin	
	ME	$MeOH:H_2O$	80:20				secologanoside; demethyloleuropein; luteolin glucoside isomer; verbascoside; oleuropein isomer; ligstroside; oleuropein	54
	ME	H_2O	100	100	30		caffeic acid; verbascoside; oleuropein; luteolin 7-*O*-glucoside; rutin; apigenin 7-*O*-glucoside and luteolin 4′-*O*-glucoside	51
Olea europaea L. mill wastewater	ME	EtOAC	100		30	defatted with *n*-hexane	hydroxytyrosol; tyrosol; *p*-coumaric acid; sinapic acid; verbascoside; hydroxytyrosol glucoside; apigenin-7-glucoside; luteolin; luteolin-7-*O*-glucoside; luteolin-7-*O*-rutinoside	49
	ME	EtOAC	100			defatted with *n*-hexane	hydroxytyrosol; gallic acid; tyrosol; *p*-coumaric acid; oleuropein aglycon	47
Olea europaea L. pomace	MMM	H_2O	100	RT	5−10	90−160 W, 20 kHz	hydroxytyrosol; comselogoside; elenolic acid derivative; tyrosol	27
	ME	H_2O	100	40	60			
	ME	$CHCl_3$	100		>500		hydroxytyrosol; gallic acid; tyrosol; oleuropein; caffeic acid; ligstroside aglycon; oleuropein aglycon; ferulic acid; vanillic acid	45
		$CHCl_3$:MeOH	90:10					
		MeOH	100					
	PLE	$EtOH:H_2O$	80:20	80	10	defatted with petroleum ether	3,4-DHPEA-EDA; oleuropein aglycon; elenolic acid; hydroxytyrosol; luteolin-7-*O*-glucoside; luteolin; ligstroside derivative	53
Allium cepa L. skin	ME	$MeOH:H_2O:HCl$	70:29.5:0.5	35	90		quercetin-4′-*O*-glucoside; quercetin-3,4′-*O*-diglucoside	59
Lycopersicon esculentum L. processing waste	ME	MeOH	100				*p*-coumaric acid; caffeic acid; chlorogenic acid; chrysin; naringenin	60
Garcinia brasiliensis Mart. leaves	percolation	MeOH + EtOAC	100				morelloflavone-7″-*O*-glucoside; gardinia biflavonoid 2a glucoside; apigenin-8-*C*-glucoside; apigenin-2″-*O*-deoxyhexoside-*C*-hexoside	102
Myrciaria dubia (Kunth) McVaugh peel	ME	$EtOH:H_2O$	80:20	RT	90		4-*O*-caffeoylquinic acid; *p*-coumaroyl hexoside; (iso)liquiritigenin-*O*-hexoside; myricetin-*O*-pentoside; myricetin-*O*-deoxyhexoside; apigenin-*O*-hexoside; ellagic acid derivatives	91
	ME	$EtOH:H_2O$ (acidified)	80:20	RT	120		delphinidin-3-*O*-glucoside; cyanidin-3-*O*-glucoside	

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Table 1. continued

agro-industrial residue	extraction technique	extraction conditions: solvent and proportion		temperature (°C)	time (min)	others	main extracted bioactive compounds	references
Myrciaria dubia (Kunth) McVaugh seed	ME	EtOH:H_2O	80:20	RT	90		valoneic acid dilactone; di-HHDP-galloyl-glucoside; ellagic acid hexoside; ellagic acid pentoside; ellagic acid; ellagic acetyl rhamnoside	91
Myrciaria dubia (Kunth) McVaugh pulp	ME	EtOH:H_2O	80:20	RT	90		*p*-coumaroyl hexoside; ferulic acid hexoside; myricetin-*O*-hexoside; myricetin-*O*-pentoside	91
	ME	EtOH:H_2O (acidified)	80:20	RT	120		cyanidin-3-*O*-glucoside	
Carissa macrocarpa (Eckl.) A.DC. leaves	ME	EtOH:H_2O	80:20	RT	120		B-type (epi)catechin derivatives; quercetin-*O*-deoxyhexoside-*O*-deoxyhexosyl-hexoside; quercetin-3-*O*-rutinoside	103
Carissa macrocarpa (Eckl.) A.DC. stems	ME	EtOH:H_2O	80:20	RT	120		*trans*-4-*O*-caffeolyquinic acid; B-type (epi)catechin derivatives; A-type (epi) catechin trimer; quercetin-3-*O*-rutinoside	103
Carissa macrocarpa (Eckl.) A.DC. flowers	ME	EtOH:H_2O	80:20	RT	120		kaempferol-3-*O*-rutinoside; quercetin-3-*O*-rutinoside; kaempferol-*O*-deoxy-hexoside-*O*-deoxyhexosyl-hexoside isomer 4; *cis*-5-*p*-coumaroylquinic acid; *cis*-4-*O*-caffeolyquinic acid	103
Actinidia deliciosa (A.Chev.) C.F.Liang and A.R.Ferguson peel	ME	EtOH:H_2O	80:20	RT	120		caffeic acid derivative; caffeic acid hexoside; dimethyl caffeic acid hexoside; epicatechin; B-type (epi)catechin derivatives; acetyl-dimethyl caffeic acid hexoside; quercetin-3-*O*-glucoside; quercetin-3-*O*-rhamnoside	104
Actinidia deliciosa (A.Chev.) C.F.Liang and A.R.Ferguson pulp	ME	EtOH:H_2O	80:20	RT	120		caffeic acid dihexoside; caffeic acid derivative; caffeic acid hexoside; dimethyl caffeic acid hexoside; epicatechin; quercetin-3-*O*-rutinoside; kaempferol-3-*O*-rutinoside; quercetin-3-*O*-rhamnoside	104
Ficus carica L. peel	ME	EtOH	0−100 (pH 3)	20−90	5−68.8		cyanidin 3-rutinoside	26
	MAE	EtOH	0−100 (pH 3)	40−115	5−35	400 W		
	UAE	EtOH	0−100 (pH 3)	30−35	5−55	100−400 W		
Persea americana Mill. peel	ME	EtOH:H_2O	80:20	RT	120		4-*O*-caffeoylquinic acid; 5-*O*-caffeoylquinic acid; B-type (epi)catechin dimer; B-type (epi)catechin trimer; B-type (epi)catechin tetramer; quercetin-dihexoside; quercetin-pentoside-hexoside	105
Persea americana Mill. kernel	ME	EtOH:H_2O	80:20	RT	120		*cis*-3-*O*-caffeoylquinic acid; *trans*-3-*O*-caffeoylquinic acid; B-type (epi)catechin dimer; *cis*-3-*p*-coumarouylquinic acid; *cis*-3-*p*-coumarouylquinic acid; catechin; epicatechin; B-type (epi)catechin trimer	105
Rubus idaeus L. leaves	ultrasonic bath	MeOH:H_2O	70:30	RT	180		ellagic acid; rutin; caffeic acid; epigallocatechin gallate; ferulic acid catechin; 5-*O*-caffeoylquinic acid	106
Rubus fruticosus L. leaves								
Myrciaria jaboticaba (Vell.) Berg pulp	ME	EtOH:H_2O	80:20	RT	120		bis-HHDP-glucose isomer; trisgalloyl-HHPD-glucose isomer; galloyl-bis-HHDP-glucose isomer; galloyl-HHDP-glucose; digalloyl-HHDP-glucose isomer; castalagin; trigalloyl-HHDP-glucose; pentagalloylglucose; quercetin-2-*O*-rhamnoside	107
		EtOH:H_2O (acidified)	80:20	RT	120		delphinidin-3-*O*-glucoside; cyanidin-3-*O*-glucoside	
Citrus limon (L.) Osbeck peel	ME	MeOH + petroleum ether + EtOAC					ferulic acid; sinapic acid; *p*-coumaric acid; ascorbic acid	57
Citrus sinensis (L.) Osbeck peel	ME	MeOH + petroleum ether + EtOAC					ferulic acid; sinapic acid; *p*-coumaric acid; ascorbic acid	57
	ME	EtOH:H_2O	80:20	35	90	−125 W	eriocitrin; narirutin; naringin; hesperidin; neohesperidin; didymin; sinensetin; 3′,4′,5,5′6,7-hexamethoxyflavone; tangeretin; nobiletin	23
	UAE	EtOH:H_2O	80:20	35	90	125 W		
	MAE	EtOH:H_2O	80:20	35	0.5	170 W		
	SC-CO_2	EtOH:H_2O	80:20	35	90	22 MPa		

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Table 1. continued

agro-industrial residue	extraction technique	extraction conditions: solvent and proportion		temperature (°C)	time (min)	others	main extracted bioactive compounds	references
Citrus paradisi Macfad. peel	ME	MeOH + petroleum ether + EtOAC					ferulic acid; sinapic acid; ascorbic acid	57
Citrus paradise Changshan-huyou (*Citrus sinensis* (L.) Osbeck × *Citrus grandis* (L.) Osbeck) peel	UAE	$MeOH:H_2O$	80:20	RT			naringin; neohesperdin; *p*-coumaric acid; ferulic acid; sinapic acid; vanillic acid	108
Mangifera indica L. seed kernel	soxhlet	MeOH	100	35	>500	0.3 bar	3-*β*-galactopyrasonyl glucose; shikimic acid; quinic acid; galloyl glucose; galloyl diglicoside; 5-*O*-galloylquinic acid; mangiferin; methyl digallate ester; anacardic acid; ginkgoic acid; tetragalloyl glucose; pentagalloyl glucose	61
Mangifera indica L. pulp	UAE	$MeOH:CH_2O_2:H_2O$ + $MeOH:H_2O$	80:2:18		30		shikimic acid-hexamalonate; shikimic acid-hexoside; gallic acid; epicatechin gallate-hexamalonate; vanillic acid; chlorogenic acid; protocatechuic acid	58
			80:20		30			
Mangifera indica L. peel	ME	MeOH	100		180	defatted with *n*-hexane	methyl gallate; gallic acid; tetra-*O*-galloyl-glucoside; penta-*O*-galloyl-glucoside; mangiferin; maclurin 3-*C*-(2-*O*-galloyl)-*β*-D-glucoside; maclurin 3-*C*-(2,3-di-*O*-galloyl)-*β*-D-glucoside; iriflophenone 3-*C*-*β*-D-glucoside; maclurin 3-*C*-*β*-D-glucoside	109
Vitis vinifera L. seed	ME	$MeOH:H_2O$ (acidified)	80:20		360		gallic acid; caffeic acid; chlorogenic acid; *p*-coumaric acid; catechin hydrate; epicatechin; epicatechin gallate	63
Vitis vinifera L. pomace	ME	$MeOH:H_2O$	80:20	RT	120		*β*-type (epi)catechin derivatives; epicatechin; gallic acid; galloyl glucose; catechin; quercetin-glucuronide; laricitrin-3-*O*-galactoside; syringetin-3-*O*-galactoside; laricitrin-3-*O*-rhamnose-7-*O*-trihydroxycinnamic acid; syringetin rutinoside derivative	66
	ME	$C_3H_6O:H_2O$	50:50	18	60		(−)-epicatechin gallate; (+)-catechin hydrate; gallic acid; quercetin; isorhamnetin; *p*-coumaric acid; malvidin 3-*O*-glucoside; pelargonidin 3-*O*-glucoside	68
		$MeOH:H_2O$	70:30	18	60		(−)-epicatechin gallate; (+)-catechin hydrate; gallic acid; quercetin; isorhamnetin; malvidin 3-*O*-glucoside; pelargonidin 3-*O*-glucoside	
		Pec-H_2O	0.01:99.9	18	60		(−)-epicatechin gallate; (+)-catechin hydrate; gallic acid	
		petroleum ether	100	18	60		(−)-epicatechin gallate; quercetin; ferulic acid; *p*-coumaric acid	
	ME	MeOH (0.1% HCl)	100	RT	>500		gallic acid; syringic acid; vanillic acid; caffeic acid; quercetin; rutin; catechin; malvidin-3-*O*-glucoside; malvidin-3-*O*-acetylglucoside; malvidin-3-*O*-*p*-coumaroylglucoside	67
Vitis vinifera L. skin	ME	$MeOH:H_2O$ (acidified)	80:20		360		caffeic acid; catechin hydrate; quercetrin hydrate; rutin hydrate; *trans*-resveratrol	63
	ME	EtOH	100	RT	180	defatted with *n*-hexane	hydroxytyrosol; gallic acid; tyrosol; protocatechinic acid; vanillic acid; caffeic acid; syringic acid; *o*-coumaric acid; *p*-coumaric acid; catechin; epicatechin; cyanidin glycosides	64

[a]Notes: pressurized liquid extraction (PLE); microwave assisted extraction (MAE); supercritical CO_2 extraction (SC-CO_2); ultrasound assisted extraction (UAE); maceration extraction (ME); multifrequency multimode modulated (MMM) ultrasonic extraction; methanol (MeOH); ethyl acetate (EtOAC); water (H_2O); chloroform ($CHCl_3$); ethanol (EtOH); acetone (C_3H_6O); diethyl ether [$(C_2H_5)_2O$]; acetic acid (CH_3COOH); formic acid (CH_2O_2); hydrochloric acid (HCl); dialdehydic form of elenolic acid linked to hydroxytyrosol (3,4-DHPEA-EDA); pectinase (Pec); room temperature (RT).

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4. BIOACTIVE COMPOUNDS IN AGRO-INDUSTRIAL RESIDUES

Agro-industrial residues have been explored as a source of natural bioactive compounds, such as flavonoids, phenolic acids (e.g., hydroxycinnamic and hydroxybenzoic acid derivatives), tannins, carotenoids, tocopherols, phytosterols, or arabinoxylans. These bioactive compounds exhibit a wide range of potential healthy properties including antiallergenic, antiatherogenic, anti-inflammatory, antimicrobial, anticarcinogenic, antithrombotic, antioxidant, cardioprotective, vasodilatory, or prebiotic effects, among others.[32,33] Numerous examples of the extraction of bioactive compounds from agro-industrial wastes can be found in the literature, as summarized in Table 1.

Phenolic compounds with antioxidant potential have been identified in agricultural residues, such as sugar cane bagasse, corn husk, peanut husk, coffee cherry husk, rice bran, or wheat bran.[34−37] Vijayalaxmi et al. (2015)[37] identified five compounds as the major bioactive compounds present in most of these residues: gallic acid, ferulic acid, epicatechin, quercetin, and kaempferol derivatives. Huang and Lai (2016)[35] established the profiles of bioactive compounds of outer and inner *O. sativa* L. bran from six colored rice samples, detecting protocatechuic, vanillic, ferulic, and *p*-coumaric acids as the most abundant phenolic acids present and α- and γ-tocopherols and γ-tocotrienol as the main vitamin E components. The major anthocyanins present in both black and red *O. sativa* L. were peonidin 3-glucoside, cyanidin 3-rutinoside, and cyanidin 3-glucoside.

A detailed work about the most abundant phenolic compounds in cork and pulp industrial residues was developed by S. A. O. Santos and co-workers.[38−41] The barks of *Eucalyptus globulus* Labill., *Eucalyptus grandis* W.Hill., *Eucalyptus urograndis* (*E. grandis* W.Hill. × *E. urophylla* S.T.Blake), and *Eucalyptus maidenii* F.Muell. as well as the cork from *Quercus suber* L. and its byproducts (cork powder and black condensates) are constituted by different types of phenolic compounds. Epicatechin and quercetin-glucuronide were revealed as the major phenolic compounds in *E. grandis* and *E. urograndis* bark, followed by ellagic acid-rhamnoside and ellagic acid in *E. grandis* and by galloyl-bis-hexahydroxydiphenoyl (HHDP)-glucose and gallic acid in *E. urograndis*. Catechin, chlorogenic acid, and methyl-ellagic acid-pentose were the major compounds in *E. maidenii* bark. Moreover, ellagic acid-rhamnoside, dihydroxy-isopropylchromone-hexoside, and dihydroxy-(methylpropyl)isopropylchromone-hexoside were referenced for the first time in these species.[41] Santos et al. (2011)[39] obtained the phenolic profile of *E. globulus* bark using different solvents (methanol, water, and methanol/water), with methanol/water mixtures being the most efficient to isolate polyphenols. Digalloylglucose was identified as the major compound in the methanol and methanol/water extracts, followed by isorhamnetin-rhamnoside in the methanol extract and by catechin in the methanol/water extract, whereas in the water extract catechin and galloyl-HHDP-glucose were identified as the predominant components. Other compounds were referenced for the first time as constituents of *E. globulus* bark, namely, quinic, dihydroxyphenylacetic, and caffeic acids, bis(hexahydroxydiphenoyl (HHDP))-glucose, galloyl-bis-(HHDP)-glucose, galloyl-HHDP-glucose, isorhamnetin-hexoside, quercetin-hexoside, methylellagic acid (EA)-pentose conjugate, myricetin-rhamnoside, isorhamnetin-rhamnoside, mearnsetin, phloridzin, mearnsetin-hexoside, luteolin, and a proanthocyanidin B-type dimer. In another work, Santos, Villaverde, and Silva et al. (2012)[40] analyzed the supercritical fluid extraction of phenolic compounds from *E. globulus* bark, using supercritical CO_2 alone and modified with water, ethyl acetate, and ethanol. In terms of the extraction yield and antioxidant activity of phenolic components, the best results were achieved with CO_2/EtOH, recovering much higher quantities of eriodictyol, naringenin, and isorhamentin than in the conventional solid/liquid extracts obtained before.

The major components identified in the cork from *Quercus suber* L. were ellagic acid, followed by gallic, protocatechuic, and caffeic acids and esculetin.[38] Those authors identified for the first time salicylic acid, eriodictyol, and quinic acid in cork extracts. Another byproduct from the cork industry is the effluent generated from the plank immersion on boiled water to improve characteristics such as elasticity and homogeneity and to make it flat. This wastewater also contains considerable amounts of phenolic compounds, namely, tannins and phenolic acids, such as gallic, protocatechuic, syringic, ferulic, and ellagic acids.[42]

From the cocoa and chocolate industry, the main byproducts obtained from the cocoa bean are pod husks, bean shells, and mucilage, which are recognized as important sources of bioactive compounds.[43] Phenolic compounds recovered from *Theobroma cacao* L. pod husk mostly consist of catechin, quercetin, epicatechin, and gallic, coumaric, and protocatechuic acids[43] and also flavonols (kaempferol) and flavones (linarin).[44]

The wastes generated in the olive oil industry are also important sources of phenolic compounds[27,45−54] with antiproliferative and antimicrobial properties.[46,48,51] Besides differences in the olive oil extraction technique, the phenolic composition of the olive wastes depends on the geographical areas, as they influence agricultural, varietal, and seasonal practices, e.g., the fruit variety and maturity stage.[46,49] Oleuropein is the most abundant compound present in *Olea europaea* L. leaves,[51,52,54,55] followed by verbascoside and isoverbascoside, oleoside, hydroxytyrosol (3,4-dihydroxyphenolethanol; 3,4-DHPEA), luteolin-7-*O*-glucoside, and ligstroside,[52] as well as apigenin-7-*O*-glucoside and luteolin-4′-*O*-glucoside.[51] Studies were performed to characterize the phenolic profile of olive wastes (mill wastewaters and olive cake) from four different areas of a region in Morocco,[48,49] concluding that the phenolic composition was different according to the geographical area, with hydroxytyrosol, tyrosol, *p*-coumaric acid, and hydroxytyrosol glucoside as the main compounds. El-Abbassi et al. (2012)[47] also reported hydroxytyrosol, gallic acid, and *p*-coumaric acid as the most abundant phenolic compounds in the olive mill wastewaters. Obied et al. (2008)[50] identified verbascoside as the most potent antioxidant present in Australian olive mill wastes, followed by 3,4-dihydroxyphenylethyl alcohol-deacetoxyelenolic acid dialdehyde (3,4-DHPEA-DEDA).

As mentioned above, the total phenolic content and composition recovered from the biomass also depend on the extraction procedure. The most interesting residue of the olive oil industry is the olive pomace, also known as olive cake. High amounts of oleuropein derivatives, such as elenolic acid (EA), the dialdehydic form of elenolic acid linked to hydroxytyrosol (3,4-DHPEA-EDA), oleuropein aglycone (3,4-DHPEA-EA), the dialdehydic form of elenolic acid linked to tyrosol (*p*-HPEA-EDA), and hydroxytyrosol, were isolated from olive

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pomaces from the region of Les Garrigues (Catalonia, Spain).[53] Cioffi et al. (2010)[45] studied the phenolic composition of virgin olive oil and olive oil pomace from the Campania region, in Italy, and found that oleuropein, oleuropein aglycone, ligstroside aglycone, hydroxytyrosol, tyrosol, and gallic acid are the most abundant phenolic compounds in both types of samples. More recently, Antónia Nunes et al. (2018)[27] compared a multifrequency multimode modulated (MMM) ultrasonic technique with a conventional solid−liquid method for the extraction of phenolic compounds from olive pomace of the Trás-os-Montes region in Portugal. The MMM technique increased the concentration of the extracted antioxidant compounds in a shorter period of time. In this sample, the major compounds identified were hydroxytyrosol > comselogoside > elenolic acid derivative > tyrosol > oleoside riboside.

Several fruits and vegetable wastes have also been reported to contain relevant amounts of phenolic compounds with high antioxidant potential. As examples, potato peel,[56] fruit peel[57] and pulp,[58] onionskin,[59] and tomato processing wastes[60] can be indicated. Gorinstein et al. (2001)[57] evaluated the antioxidant properties of some citrus fruit wastes, namely, *Citrus limon* (L.) Osbeck, *Citrus sinensis* (L.) Osbeck, and *Citrus paradise* Macfad. peels, reporting high contents of ferulic, sinapic, *p*-coumaric, caffeic, and ascorbic acids. Concerning onion wastes, Benítez et al. (2011)[59] observed that brown skin, outer and inner scales and top and bottom of *Allium cepa* L. were particularly rich in quercetin-4′-*O*-glucoside and quercetin-3,4′-*O*-diglucoside. In *Lycopersicon esculentum* L. processing wastes, mainly consisting of skin and seeds, the major compounds detected were *p*-coumaric, caffeic, and chlorogenic acids as phenolic acids and chrysin and naringenin as flavonoids.[60] Castro-Vargas et al. (2019)[61] explored the potential of the agro-industrial waste from two Colombian mango cultivars, "sugar mango" and "Tommy Atkins", as sources of bioactive phenolic compounds. The "sugar mango" kernel extracts presented the highest bioactive properties, with mangiferin and several galloyl glucosides as the most abundant compounds. Chlorogenic, gallic, vanillic, and protocatechuic acids were the major phenolic compounds found in *Mangifera indica* L. "Ataulfo" pulp,[58] which tended to increase with fruit ripening and also their contribution for antioxidant activity. More recently, Coelho et al. (2019)[62] demonstrated that mango peels could be used to produce liqueurs by alcoholic maceration. These liqueurs were revealed to have high antioxidant activity, and quantitatively, the main bioactive compounds found were flavanols (epicatechin gallate, epigallocatechin gallate), flavonols (quercetin-3-*O*-glucoside and rutin), and phenolic acids (gallic acid, *o*-coumaric acid, and syringic acid).

Wine production generates different wastes, such as stem, skin, and seeds[63,64] and pomaces.[65−68] Ribeiro et al. (2015)[67] reported the presence of high amounts of gallic, syringic, vanillic, and caffeic acids, quercetin, rutin, and catechin, in addition to anthocyanins, especially malvidin-3-*O*-glucoside, malvidin-3-*O*-acetylglucoside, and malvidin-3-*O*-*p*-coumaroyl-glucoside, in *Vitis vinifera* L. pomace. Doshi et al. (2015)[63] suggested that grape seeds, skin, and stem extracts not only have high antioxidant properties but also have the potential as insulin secretagogues, which might be useful in the treatment of type II diabetes. Gallic acid, catechin, and epicatechin were identified as major phenolic compounds in winery wastes,[64] but hydroxytyrosol, tyrosol, cyanidin glycosides, and various phenolic acids, such as caffeic, syringic, vanillic, *p*-coumaric, and *o*-coumaric acids, were also found.

5. IONIZING RADIATION TECHNOLOGIES

Radiation technologies offer versatile tools that play an important role in supporting sustainable development.[69] Ionizing radiation does not require the addition of chemicals or heat to induce changes in the matter, having a much lower carbon footprint than other technologies. In this way, it is more cost-effective and environmentally friendly than traditional alternatives, requiring less energy and generating less waste.[70]

There are different sources of ionizing radiation used in different sectors of industry and agriculture:[71,72] γ-radiation, that is constituted by photons produced from radioactive isotopes cobalt-60 (^{60}Co) and cesium-137 (^{137}Cs), and accelerators, capable of producing electron beams and X-rays. Radioisotopes are useful due to their permanent source, high efficiency associated with their penetrating power and their easy replacement. Electron-beam accelerators do not rely on a radioactive source and present the advantage that they are able to be switched on and off depending on the necessity.

Radiation technologies are safe processes becoming part of the solution for many countries to develop new tools and approaches in different areas, ranging from health and the environment to industry and infrastructure.[69]

5.1. Radiation Processing. Ionizing radiation is currently used for different applications: medical device sterilization,[73] material modification,[74] heritage preservation,[75] or wastewater treatment,[76] among others. Contrarily to ethylene oxide treatment that can leave toxic residues on treated products, ionizing radiation can be safely used to sterilize healthcare products such as syringes, surgical gloves, or tissue allografts in sealed packages, in order to reduce the microbiological contamination to acceptable levels.[77] This technology can also be applied on the preparation and functionalization of hybrid materials that are used for biomedical applications and food packaging.[74] On the other hand, Borrely et al. (2016)[76] demonstrated that electron-beam radiation was capable of reducing the color (>90% at 2.5 kGy) and toxicity of three different textile effluents and Madureira et al.[78] showed the radiolytic degradation of phenolic acids present in wastewaters from the cork industry. In addition, γ-radiation was used to decontaminate and preserve parchment documents as an alternative to the chemical (e.g., fumigants) and nonchemical (e.g., modified indoor atmospheres) treatments that induce toxicity and deterioration of the documents.[75]

Both types of irradiation have also been used for food processing and have shown to be effective in reducing pathogenic bacteria, eliminating parasites, decreasing post-harvest sprouting, and extending the shelf life of fresh perishable food.[70] Furthermore, irradiation can also be applied to enhance extraction or improve the bioactive properties of some of the chemical compounds present in food, wastes, and plants. In the present review, we will focus on the extractability and bioactive properties of agro-industrial residues.

5.2. Ionizing Radiation in Agro-Industrial Residues: Enhancement of Phenolic Compounds Extractability and Bioactive Properties. Although there is no ideal method for extracting bioactive compounds, ionizing radiation technologies have proven to improve the extraction yield and bioactive properties of agro-industrial residues. In recent years, several studies have been published regarding the extractability

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Table 2. Irradiation Effects on Phenolic Compounds and Bioactivity of Agro-Industrial Residues[a]

agro-industrial residue	extract	radiation source	doses	main results	identified compounds	references
Quercus suber L. wastewater		γ-radiation	20, 50, and 100 kGy	increase of DPPH scavenging activity in 34% at 100 kGy; increase of reducing power in 33% at 100 kGy; increase of β-carotene bleaching inhibition in 62% at 100 kGy	n.i.	79
Curcuma alismatifolia Gagnep. leaves	80% methanol	γ-radiation	10, 15, and 20 Gy	increase of total phenolics, total flavonoids, and antioxidant activity with increasing radiation doses	salicylic acid; caffeic acid; catechin; epicatechin; cinnamic acid; ellagic acid; resorcinol; rutin; naringin; quercetin; myricetin	80
Rosmarinus officinalis L. leaves	methanol; ethanol; water	γ-radiation	30 kGy	increase of the antioxidant activity of ethanol and water extracts with irradiation; increase on the total phenolic content in water extracts with irradiation	n.i.	81
Ziziphus mauritiana Lam. leaves	methanol; *n*-hexane; water	γ-radiation	2.5, 5, 7.5, 10, and 12.5 kGy	enhancement of certain phytochemicals (phenolics, flavonoids, tannins, and saponins) at doses up to 12.5 kGy; increase of DPPH scavenging activity and extraction yields of *Ziziphus mauritiana* Lam. leaf extracts with irradiation	n.i.	82
Prunus amygdalus Batsch skin	40% ethanol	γ-radiation	2−16 kGy	Blue Diamond company: increase in phenolic content at irradiation doses higher than 4.05 kGy; Campos Brothers company: increase in phenolic content at irradiation doses higher than 12.7 kGy	n.i.	83
apple pomace	84.5% methanol + 65% acetone	γ-radiation	1 and 2 kGy	more extractable individual phenolic compounds at 1 kGy; increase of total phenolic compounds and antioxidant potential at 1 kGy	quercetin-3-D-galactoside; quercetin-3-β-D-glucoside; quercetin-3-*O*-rhamnoside; hydroxycinnamic acid; chlorogenic acid; dihydrochalcones; phloridzin; epicatechin; epicatechin; quercetin-3-*O*-rutinoside; procyanindin B2; procyanindin B1; catechin	84
Castanea sativa Mill. skin	methanol	γ-radiation	0.27 and 0.54 kGy	enhancement of antioxidant potential of skins at 0.54 kGy	n.i.	85

[a]n.i.: not identified.

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and bioactive properties improvement of chemical compounds from irradiated food and agro-wastes, demonstrating the high potentiality of this treatment (Table 2).

A study developed by Madureira et al. (2017)[79] revealed that γ-radiation at 100 kGy was capable of improving (34−62%) the antioxidant activity of wastewaters generated during *Q. suber* L. processing. Taheri, Abdullah, Karimi, Oskoueian, and Ebrahimi (2014)[80] demonstrated that 20 Gy of γ-radiation was enough to significantly increase the total phenolics (2-fold) and total flavonoids (2-fold) contents in *Curcuma alismatifolia* Gagnep. leaves compared to the nonirradiated ones. An increase of 35% on total phenolic content was observed in water extracts of *Rosmarinus officinalis* L. leaves irradiated at 30 kGy but not in methanol or ethanol extracts.[81] The authors associated this increase with the presence of diterpenes in rosemary that could result in water-soluble quinone-type compounds by γ-radiation. For their part, Khattak and Rahman (2016)[82] showed that γ-radiation doses up to 12.5 kGy enhanced the levels of certain phytochemicals (phenolic acids, flavonoids, tannins, and saponins) and increased the DPPH scavenging activity and extraction yield of *Ziziphus mauritiana* Lam. leaves extracts.

Harrison and Were (2007)[83] reported two different irradiation levels causing the enhancement of the total phenolic content and antioxidant activity of *Prunus amygdalus* Batsch. skin extracts, although dependent on the almond skin origin. Almond skins supplied by the company Blue Diamond Growers showed an increase in phenolic content at irradiation doses higher than 4.05 kGy (about 45% compared to the control), while almond skins from Campos Brothers showed an increase in phenolic content at irradiation doses higher than 12.7 kGy (about 20% compared to the control).

Apple pomace is another interesting source of phenolic compounds. Irradiation of apple pomace flour at 1 kGy enhanced the extractability of individual and total phenolic compounds, as well as the antioxidant potential.[84] More specifically, total phenolic compounds increased from 563 ± 50 mg/100 g (0 kGy) to 661 ± 33 mg/100 g (1 kGy) and antioxidant capacity by FRAP assay increased more than 50% (from 36.19 ± 1.36 to 57 ± 1 mmol trolox equivalents/g for nonirradiated and irradiated at 1 kGy sample, respectively).

Chestnuts are also another important source of polyphenols with antioxidant potential. António et al. (2011)[85] reported an increase of the antioxidant capacity in *Castanea sativa* Mill. skins irradiated at 0.54 kGy of 64% evaluated by the β-carotene bleaching inhibition assay.

5.3. Radiation Chemistry. The effect of ionizing radiation on extraction yields can vary depending on the compounds that constitute the studied matrixes. The increase in total phenolic and flavonoid contents on irradiated samples could be due to the release of these compounds from matrix structures, increasing the extractability of certain molecules, or the degradation of larger compounds (e.g., tannins) into smaller ones by the radiolytic action of ionizing radiation.[86] The higher extractability can be explained by changes in cellular structures, namely, by the depolymerization and dissolution of the cell wall polysaccharides by irradiation.[83,87,88] Moreover, irradiation is recognized as promoting the activity of phenylalanine ammonia-lyase, responsible for the synthesis of phenolic compounds,[89] and, consequently, the enhancement of antioxidant activity.

In addition, as mentioned above, the increase or decrease on the antioxidants is strongly dependent on the solvents used for the extraction.[81,82,90]

6. APPLICATIONS OF BIOACTIVE COMPOUNDS EXTRACTED FROM AGRO-INDUSTRIAL RESIDUES

With the growing interest of consumers toward food bioactives that provide beneficial effects on human health promotion and disease risk reduction, the extracts from irradiated residues with improved properties can be incorporated in food products and contribute for the development of functional foods. Different investigations have focused on the development of functional foods through fortification, which consists of incorporating one or more bioactive compounds in order to correct or improve a potential biological activity of the food product. There are several studies concerning the incorporation of bioactive compounds from agro-industrial residues into different dietary matrixes of high importance, namely, bread, pastry, biscuits, and yogurt.[91−94] *Myrciaria dubia* (Kunth) McVaugh peel was used to fortify yogurt,[91] resulting in an enhancement in bioactive components in the new product without significantly altering its nutritional composition and fatty acid profile. *Punica granatum* L. epicarp extracts were incorporated in a typical Brazilian pastry product.[92] The new formulations showed greater antioxidant activity that remained unchanged during the 14 days of storage. At the same time, the cake prepared by new formulations presented a rose color and a soft texture, which can also be important factors for the consumer acceptance. Also, Hallabo et al.[93] substituted wheat flour of biscuits by 6% peel powders of *M. indica* L. cv. Copania, *A. cepa*, and *Solanum tuberosum* L. The obtained results demonstrated that the new prepared biscuits had higher antioxidant activity, total carotenoids, and total polyphenols than the control ones. In another study performed by Mildner-Szkudlarz et al.,[94] a new formulation for sourdough mixed rye bread was produced incorporating a maximum of 6% grape byproducts and presented an improvement in antioxidant activity due to the higher values of phenolic compounds.

There are a few patents on the application of agro-industrial residues on food products.[95] For example, a straightforward method for obtaining polyphenol extracts from white grape residues to having antioxidant and antibacterial properties was patented to be used on an industrial scale in the cosmetic, pharmaceutical, and/or food industries.[96,97] Another process that utilizes cheese/yogurt whey and fruit pomace to directly yield extruded, ready-to-eat products such as breakfast cereals, healthy snacks, protein puffs, and nutrition bars, among other food items, was also patented.[98]

The actual trend is the bioactive compounds production by extraction from residual food sources,[95] although further research on the irradiation of different agro-industrial residues should be performed to increase its applicability and to valorize the wastes by enhancing their health benefits and maintain environmental sustainability.

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Notes

The authors declare no competing financial interest.

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