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REVIEW

Recent trends and advancements in microbial tannase-catalyzed biotransformation of tannins: a review

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Abstract

The outburst of green biotechnology has facilitated a substantial upsurge in the usage of enzymes in a plethora of industrial bioconversion processes. The tremendous biocatalytic potential of industrial enzymes provides an upper edge over chemical technologies in terms of safety, reusability, and better process control. Tannase is one such enzyme loaded with huge potential for bioconversion of hydrolysable tannins to gallic acid. Tannins invariably occur in pteridophytes, gymnosperms, and angiosperms and predominately cumulate in plant parts like fruits, bark, roots, and leaves. Furthermore, toxic tannery effluents from various tanneries are loaded with significant levels of tannins in the form of tannic acid. Tannase can be principally employed for debasing the tannins that predominately occur in the toxic tannery effluents thus providing a relatively much cheaper measure for their biodegradation. Over the years, microbial tannase-catalyzed tannin degradation has gained momentum. The plentious availability of tannin-containing agro- and industrial waste paves a way for efficient utilization of microbial tannase for tannin degradation eventually resulting into gallic acid production. Gallic acid has received a great deal of attention as a molecule of enormous therapeutic and indusrial potential. The current worldwide demand of gallic acid is 8000 t per annum. As a matter of fact, bioconversion of tannins into gallic acid through fermentation has not been exploited completely. This necessitates further studies for development of more efficient, economical, productive processes and improved strains for gallic acid production so as to meet its current demand.

Keywords Bioconversion · Tannase · Gallic acid · Hydrolysable tannins · Bioremediation

Introduction

Biotransformation process principally utilizes biocatalysts either as whole cells or as enzyme(s) for the manufacturing of broad range of bio products. Commercially, the biotransformation process offers various advantages over chemical technologies in terms of better control over various process parameters, capability to genetically alter microorganisms, better yields, credibility, safety, and reusability. As a matter of fact, their biochemical heterogeneity, capability to undergo genetic modification, and production on a higher scale in a relatively short time span through fermentation render microorganisms as an alternate approach for the enzyme production (Bharathiraja et al. 2017). A significant number of biotransformation operations have efficiently utilized several biocatalysts like cellulase, xylanase, amylase, and lipase for producing valuable products of commercial importance (Willke and Worlop 2004). Green biotechnology incites the utilization of enzymes in producing a vast number of products of industrial significance more precisely using renewable sources. A great majority of industrial enzymes used in present day era are of microbial origin since they generally exhibit relatively escalated activities in comparison to enzymes having plant and animal origin thus exemplifying an alternate source of enzymes. According to an estimate, worldwide market for industrial enzymes was evaluated around $4.2 billion in 2014 and is anticipated to hit approximately $ 6.2 billion mark globally at a compound yearly growth rate of around 7% during 2015 to 2020 (2015 a, b: Industrial Enzyme Market). A substantial proportion of industrial enzymes (around 65%) are “hydrolases” (Johannes and Zhao 2006). Tannase (E.C.3.1.1.20) being hydrolase catalyzes the biotransformation of hydrolysable tannins to simple

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phenolic molecules like gallic acid. Tannins are high molecular weight (500–3000 kDa), polyphenolic compoumds occurring in plants and are one of the major constituents of effluents released by tanning industries which are toxic to plants, animals and microorganisms, posing potential threat to both human health as well as the environment (Van de Lagemaat and Pyle 2001). Microbial tannase enzyme is loaded with tremendous potential for bioconversion of hydrolysable tannins to gallic acid. Gallic acid, the major hydrolytic product of tannic acid, has extended its utilization as a highly valuable therapeutic agent: as an antimicrobial, antiviral, antitumor, and radio protective agent as well as a potential drug (Zeida et al. 1998; Mukherjee and Banerjee 2003; Das et al. 2006; Purohit et al. 2006; Hsu et al. 2007; Aithal and Belur 2013) (Fig. 1).

Further, its utilization in agriculture sector in protecting crops and food storage renders gallic acid a molecule of impeccable commercial utility. The extremely salutary applications of gallic acid may make it a trendsetter in therapeutics as well as industries. The worldwide annual demand of gallic acid is 8000 t. China is the leading producer of gallic acid. The existing technology for gallic acid production involving the acidic hydrolysis of tannins for gallic acid production is costlier and generates high levels of toxic effluents that pose several environmental hazards. It also involves high production cost, low yield and less purity of Gallic acid (Paranthaman et al. 2009). The bioconversion of tannins into gallic acid through microbial fermentation has not been utilized completely. Evidently, the original studies on using the substantially high concentrations of tannic acid for tannase production as well as for galic acid production at fermenter level are scanty in literature. The biocatalyst tannase utilized for biotransformation of tannins to gallic acid itself holds remarkable importance at industrial level credited to its stupendous catalytic potential. The commercial level production of tannase is being undertaken by certain producers worldwide like Kikkoman (Japan), Novo Nordisk (Denmark), ASA special enzyme GmbH (Germany), Amano (Japan), Biocon (India), Julich (Germany), Wako Pure Chemical Industries, Ltd. (Japan), and Sigma–Aldrich Co. (USA). Despite the commercial utility of tannase and gallic acid in plethora of applications, there are very few research studies available in literature on their production at fermenter level. Thus, on the account of plenteous tannin containing agrowaste and industrial waste, astounding commercial potential of gallic acid, bioremediation need for the tannin waste, and exorbitant approaches for gallic acid production, a cheaper and eco-friendly green technology can be efficiently utilized to meet the current demand of gallic acid.

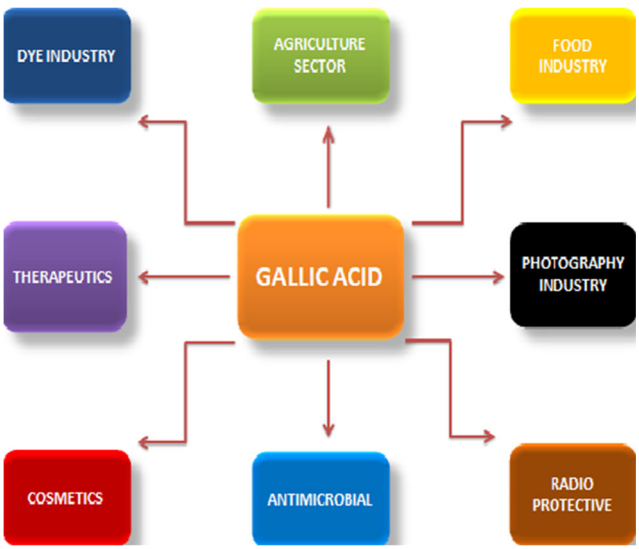

Fig. 1 Versatile applications of Gallic acid

Several reviews have been written with major emphasis on tannase production from various microbial sources, its purification, characterization, and immobilization etc. (Jana et al. 2014; Chavez Gonzalez et al. 2017). However, a comprehensive review on biocatalytic potential of microbial tannase in biotransformation of tannins to gallic acid production has not been attempted till date. Therefore, in the present review, a major emphasis has been laid on providing descriptive information on various sources of microbial tannase, statistical modeling for efficient tannase production, cloning and expression of tannase, range of tannin rich substrates for gallic acid production, tannins as pollutants, microbial transformation of tannins to gallic acid, role of various matrices in tannase immobilization for enhanced tannin hydrolysis and scientific perspectives as well as the challenges that need to be addressed for enhanced tannin hydrolysis and gallic acid production.

Tannin acyl hydrolase (TAH)

Tannase enzyme is one of the most adept biocatalysts and plays a key role in a broad range of bioconversion reactions under protein-precipitating conditions. Tannase strictly works on ester and depside bonds in hydrolysable tannins, principally gallotannins thereafter giving off gallic acid and glucose molecule (Belur and Mugeraya 2011; Yao et al. 2014). Industrial biotransformation of tannic acid to gallic acid is generally executed with tannase.

Microbial tannase sources

The most significant approach of obtaining the enzymes is via microbial way, since the microbial enzymes exhibit better stability in comparison to enzymes from other possible sources (Jana et al. 2014). More importantly, microbes can consistently produce higher titers of enzymes. Moreover, microorganisms can produce tannase in high quantities in a constant way.

Microbes can be genetically manipulated thus resulting in increased tannase activity titers (Aguilar and Gutierrez-Sanchez 2001). There are many microorganisms used for tannase production. Majority of tannase-producing organisms documented till date belong to bacteria, fungi, and yeast.

Tannase from bacteria

Numbers of bacteria have been reported to produce tannase. Most of the reported bacterial tannase producers correspond to extracellular tannase (Chavez Gonzalez et al. 2017). Over the last 25 years, the interest in bacterial tannases has risen owing to their widespread applications, ability to undergo genetic manipulations, and capability to live under extreme temperature conditions. Thus, in this regard, several tannase-producing bacteria have been identified till date among which bacterial strains belonging to genera such as *Lonepinella* (Goel et al. 2007), *Staphylococcus* (Noguchi et al. (2007), *Lactobacillus* (Guzman-Lopez et al. (2009), *Pseudomonas* (Selwal et al. 2010), *Serratia* (Belur et al. (2010), *Bacillus* (Raghuwanshi et al. (Raghuwanshi et al. 2011; Muhammad et al. 2016), *Azobacter* (Gauri et al. (2012), *Klebsiella* (Sivashanmugam and Jayaraman 2013), *Citrobacter* (Wilson et al. (2009) *Pantonea* (Pepi et al. 2010), and *Enterobacter* (Mandal and Ghosh 2013) are predominant. The molecular weights of tannase of bacterial origin usually lie within 46.5–90 kDa (Jana et al. 2013). Deschamps et al. 1983 firstly reported the extracellular production of tannase from *Bacillus*, *Corynebacterium*, and *Klebsiella* strains with the concomitant release of gallic acid and glucose.

Yeast

Only certain variants of yeasts have been documented to depolymerize tannins. (Aoki et al. 1976) documented the depolymerisation of tannins by *Candida* sp. Apart from that, only a few types of yeast have been reported with the ability to degrade tannins.

Fungi

Hadi et al. (1994) reported the ability of filamentous fungi to depolymerise tannins. As many as 120 fungal species have been documented till date as tannase producers. Majority (around 70) of them belong to genera *Aspergillus* and *Penicillium* (Chavez Gonzalez et al. 2017). The filamentous fungi pertaining to genera *Aspergillus* have been primarily utilized for tannase production (Banerjee et al. 2001). Fungi like *Aspergillus oryzae* (Bradoo et al. 1996), *Aspergillus awamori* (Beena et al. 2010), *Aspergillus fumigates* (Batra and Saxena 2005), *Aspergillus ruber* (Kumar et al. 2007), *Penicillium chrysogenum* (Bradoo et al. 1996), *Penicillium glabrum* (Van de Lagemaat and Pyle 2005), *Trichoderma viride*, and *Trichoderma hamatum* (Bradoo et al. 1996) have been reported as efficient tannase producers. The tannase-producing organisms and biochemical properties of some of the characterized tannases are listed in Tables 1 and 2.

Microbial tannase production methodologies

Submerged fermentation has been reported as immensely favored approach for production of tannase globally. Depite this, certain research invesigations involving production of tannase via solid state fermentation approach have also been reported. Selecting a proficient production approach relies on various attributes like strain to be utilized during production process, nutrient accessibility, variety, and quality of substrate being utilized.

Submerged fermentation

Submerged fermentation basically utilizes a high oxygen concentrated liquid nutrient medium for culturing the microbes. SMF has been the principally utilized approach for producing tannase as well as other enzymes at industrial level (Chavez-Gonzalez et al. 2012). Submerged fermentation is always preferred for microbial tannase production because it offers uniform fermentation conditions like substrate concentration, inducer concentration, temperature, pH, dissolved oxygen concentration, agitation, aeration, superior process control, ease in extracting the enzyme, better utilization of substrate, relatively shorter incubation time span, proficient mass, and heat transfer as well better feasiblity of statistical and kinetic modeling of the process (Rao 2010; Prasad et al. 2012). Enzyme producers usually produce enzymes using submerged fermentation with important titers in the range of grams per liter (Aguilar et al. 2007). Tannic acid serves as the carbon source as well as an inducer for production of tannase. Thus, in this context, its concentration plays a crucial role for the growth of microbes as well as production of tannase (Rao et al. 2008; Chavez-Gonzalez et al. 2012).

Das et al. (2006) utilized tannins from eight unalike plant extracts for production of tannase with *Bacillus licheniformis* KBR6 in SMF and recorded a higher activity with the tannin of *Anacardium occidentale.* Selwal et al. (2010) studied the production of tannase enzyme using *Pseudomonas aeruginosa* IIIB 8914 under submerged fermentation with the leaves of *Phylanthus emblica* (amla), *Acacia nilotica* (keekar), *Eugenia cuspidate* (Jamoa), and *Syzygium cumini* (Jamun) as substrates and reported a maximum tannase yield. Kannan et al. (2011) reported maximal enzyme activity (5.22 U/ml) of tannase from *Lactobacillus plantarum* MTCC 1407 under submerged fermentation.

SMF has been most preffered approach for bacterial tannase production with higher enzyme titers (Belur and

Table 1 Microorganisms used for the production of tannin acyl hydrolase (tannase)

Microorganism Bacteria	Reference
Achromobacter sp.	Lewis and Starkey (1969)
Bacillus pumilus	Deschamps et al. (1983)
Bacillus polymyxa	Deschamps et al. (1983)
Corynebacterium sp.	Deschamps et al. (1983)
Klebsiella planticola	Deschamps et al. (1983)
Klebsiella pneumoniae	Deschamps et al. (1983)
Paenibacillus polymyxa	Deschamps et al. (1983)
Pseudomonas solanacearum	Deschamps and Lebeault (1984)
Lonepinella koalarum	Osawa et al. (1995)
Citrobacter freundii	Kumar et al. (1999)
Bacillus licheniformis	Mondal et al. (2000)
Lactobacillus plantarum	Osawa et al. (2000)
Lactobacillus paraplantarum	Osawa et al. (2000)
Lactobacillus pentosus	Osawa et al. (2000)
Bacillus cereus KBR9	Mondal et al. 2001
Lactobacillus plantarum	Ayed and Hamdi (2002)
Pseudomonas citronellolis	Chowdhury et al. (2004)
Streptococcus bovis	Belmares et al. (2004)
Streptococcus gallolyticus	Sasaki et al. (2005)
Bacillus licheniformis KBR 6	Das et al. (2006)
Staphylococcus lugdunensis	Noguchi et al. (2007)
Lactobacillus plantarum	Kostinek et al. (2007)
Lactobacillus plantarum	Iwamoto et al. 2008
Lactobacillus buchneri	Guzman-Lopez et al. (2009)
Enterobacter cloacae	Beniwal et al. (2010)
Pseudomonas aeruginosa	Selwal et al. (2010)
Serratia ficaria DTC	Belur et al. (2010)
Bacillus sphaericus	Raghuwanshi et al. (2011)
Bacillus massieliensis	Belur et al. (2012)
Gluconacetobacter hansenii	Rani and Appaiah (2012)
Enterobacter asburiae	Mandal and Ghosh (2013)
Klebsiella pneumonia	Sivashanmugam and Jayaraman (2013)
Lactobacillus plantarum CIR1	Aguilar-Zarate et al. (2014)
Erwinia carotovora	Muslim et al. (2015)
Bacillus subtilis	Muhammad et al. (2016)
Bacillus gotthelii M2S2	Subbulaxmi and Murty (2016)
Bacillus megaterium	Tripathi and Sharma (2016)
Klebsiella pneumonia	Tahmourespour et al. (2016)
Escherichia coli	Thakur and Nath (2017a)
Fusobacterium nucleatum subs. *polymorphum* ($TanB_{Fnp}$)	Tomas-Cortazar et al. (2018)
Streptomyces sp.	Roy et al. (2018)
Yeast	
Candida sp.	Aoki et al. (1976)
Debaromyces hansenii	Deschamps et al. (1983)
Pichia spp.	Deschamps and Lebeault (1984)
Debaryomyces hansenii	Deschamps and Lebeault (1984)
Mycotorula japonica	Belmares et al. (2004)
Saccharomyces cerevisiae	Zhong et al. (2004)

Table 1 (continued)

Microorganism Bacteria	Reference
Fungi	
Aspergillus niger	Knudson 1913
Aspergillus oryzae	Iibuchi et al. (1967)
Aspergillus flavus	Yamada et al. (1968)
Aspergillus japonicus	Bradoo et al. (1996)
Penicillium notatum	Ganga et al. (1977)
Cryphonectria parasitica	Farias et al. (1994)
Rhizopus oryzae	Hadi et al. (1994)
Fusarium solani	Bajpai and Patil (1996)
Fusarium solani	Bradoo et al. (1996)
Aspergillus oryzae	Bradoo et al. (1996)
Aspergillus gallonyces	Belmares et al. (2004)
Fusarium solani	Belmares et al. (2004)
Aspergillus acolumaris	Batra and Saxena (2005)
P. glabrum	Van de Lagemaat and Pyle (2005)
P. crustosum	Batra and Saxena (2005)
R. oryzae	Mukherjee and Banerjee (2006)
A. aculeatus	Banerjee et al. (2007)
A. fumigatus	Manjit et al. (2008)
Penicillium variable	Sharma et al. (2008)
A. flavus	Paranthaman et al. (2009); Kuppusamy et al. (2014)
A. tamari	Enemour and Odibo (2009)
A. awamori	Beena et al. (2010)
A. oryzae	Abdel-Nabey et al. (2011)
vA. japonicas	Abdel-Nabey et al. (2011)
Trichoderma harzianum	Iqbal and Kapoor (2012)
A. ochraceus	Goncalves et al. (2012)
P. purpurogenum	Reddy and Rathod (2012)
Aspergillus niger	Ahmed and Rhman (2014)
A. aculeatus	Bagga et al. (2015)
Aspergillus melleus	Liu et al. (2016)
A. fumigatus CAS21	Cavalcanti et al. (2017)
A. tamarii URM 7115	de Sena et al. (2018)

Mugeraya 2011). A great majority of lab-scale tannase production from bacteria is done within 250-ml Erlenmeyer flask (Das et al. 2006). However, some research investigations involving bacterial tannase production have been carried out on fermenter scale (Raghuwanshi et al. 2011). Research investigations have been conducted with major emphasis on exploring the bacterial strains with the potential of producing higher enzyme titers. Maximal tannase production (16.54 U/mL) was recorded by Raghuwanshi et al. 2011 from *Bacillus sphaericus* with a 30 L fermenter.

Submerged fermentation approach has also been utilized for tannase production from fungi. Bajpai and Patil (1997) reported production of tannase from *Aspergillus* spp. at relatively high aeration rates. Sharma et al. (2007) studied the effect of various parameters like rate of agitation, incubation time period, sodium nitrate, and effect of tannic acid concentration on tannase production using *Aspergillus niger* in submerged fermentation. Murugan et al. (2007) reported production of tannase via SMF using *Aspergillus niger*, *Aspergillus xavus, Penicillium* spp., *Trichoderma* spp. etc. *A. niger* was reported as the most proficient tannase producer (16.77 U/mL) among all. Paranthaman et al. (2009) also utilized SMF strategy for obtaining maximum production of tannase from *A. flavus*

Table 2 Biochemical properties of some characterized tannases

Microorganism source	Substrate	pH_{Opt}	$Temp_{Opt\ (°C)}$	pI	$K_{m\ (mM)}$	V_{max}	pH stability	Temp stability (°C)	Reference
S. ruminantium	Methyl gallate	7.0	30–40	6.5–8.0	1.6	6.3 μmol/min·mg	–		Skene and Brooker (1995)
L. plantarum	Methyl gallate	8.0	40	–	0.62	–	7.5–9.0	≤ 45	Iwamoto et al. (2008)
B. sphaericus	Tannic acid	5.0	50	–	–	–	3.0–8.0	30–80	Raghuwanshi et al. (2011)
Enterobacter sp	Methyl gallate	5.5	40	–	3.7	0.166 U			Sharma and John (2011)
B. subtilis	Tannic acid	5.0	40	4.4	0.445	125.8 mM/mg/min	3.0–8.0	30–70	Jana et al. (2013)
E. cloacae	Tannic acid	5.5	50		0.00337 M	3.401 U/ ml	–	> 50	Beniwal et al. (2013)
Aspergillus niger LCF8	Tannic acid	6.0	35	4.3	–	–	3.5–8.0	≤ 45	Barthomeuf et al. (1994)
Aspergillus niger van Tieghem	Methyl gallate	6.0	60	–	0.2	5 μmol/min·mg	4.5–7.5	≤ 60	Sharma et al. (1999)
Aspergillus oryzae	Tannic acid	5.5	40	–	7.35	83 μmol/min·mg	4.5–6.0	≤ 45	Abdel-Nabey et al. (1999)
Verticillium sp. (TAH1)	Tannic acid	5.5	25	5.8	1.05	–	4.5–6.0	≤ 30	Kasieczka-Burnecka et al. (2007)
Verticillium sp. (TAH2)	Tannic acid	5.5	20	6.2	1.05	–	5.0–7.5	≤ 30	Kasieczka-Burnecka et al. (2007)
Paecilomyces variotii	Tannic acid	5.5	55	–	6.1×10^{-4}	5.6 μmol/min·mg	4.5–6.5	≤ 55	Battestin and Macedo (2007)
Arxula adeninivorans	Methyl gallate	6.0	40	–	4.4	–	5.0–7.0	≤ 50	Boer et al. (2009)
Aspergillus niger GH1	Methyl gallate	6.0	60	3.5	0.041	11.03 μmol/min·mg	4.0–6.0	≤ 50	Gomez et al. (2009)

under optimal conditions of temperature, incubation time period and substrate concentration. Srivastava and Kar (2009) obtained maximal tannase and gallic acid production from *A. niger* by utilizing pomegranate rind powder (4% *w*/*v*) as substrate under optimized conditions. Beniwal et al. (2010) utilized response surface methodology (RSM) for optimizing the process parameters for maximal production of tannase from *Aspergillus awamori* MTCC 9299.

Darah et al. (2011) reported maximal tannase production (2.81 U/ml) from *Aspergillus niger* FETL FT3 via SMF strategy. Iqbal and Kapoor (2012) studied the production of tannase by *Trichoderma harzianum* MTCC 10841 under submerged fermentation using various tannin rich materials as carbon sources. Ahmed and Rhman (2014) also documented production of tannase from *Aspergillus niger* under optimal condtions of temperature, pH, and incubation time period. Varadharajan et al. (2015) reported pomegranate rind extract as the most proficient substrate for producing tannase from *A. oryzae* by SMF approach.

Solid-state fermentation

Solid-state fermentation (SSF) has witnessed a continuous rise in its use for tannase as well as other industrial enzymes production. Solid-state fermentation (SSF) is a process that preferentially occurs in near or absolute absence of free flowing water thus employing either a natural support or an inert support as a solid material. However, the substrate must be having sufficient moisture to sustain the growth microbial growth and metabolism (Pandey et al. 2000). SSF approach has been utilized to a relatively lesser extent for production of tannase as compared to submerged fermentation (Jana et al. 2013). The research investigations conducted in recent years have claimed enhanced tannase production and better stability in accordance with pH and temperature deviations. However, the majority of literature suggests suitability of SSF for fungal tannase production by utilizing natural tannin containg agro residues as they imitate the natural conditions indispensable for fungal growth. Several natural tannin containing substrates like wheat bran, coffee pulp and tea residue, tamarind seed powder, and rice bran have been efficiently utilized for maximal tannase production via SSF. Polyurethane foam has been the most commonly used natural support amongst various other supports (Rodrıguez-Duran et al. 2011); Wilson et al. (2009) and Jana et al. (2013) documented as high as 45 times and 7 times enhanced bacterial tannase production via SSF in comparison to SMF. However the original research studies utilizing SSF for bacterial tannase production are scanty.

Various natural tannin-containing substrates jamun leaves, amla leaves (Kumar et al. 2007; Selwal et al. 2011), tamarind seed powder, baggase, ground nut oil cake, wheat bran and rice bran (Natarajan and Rajendran 2012), coffee pulp, and tea residue (Sharma et al. 2014; Bhoite and Murthy 2015) have been utilized as substrates for tannase production under SSF.

Aguilar et al. (2001) reported 2 times higher biomass yield in solid-state fermentation during the production of tannase using *Aspergillus niger* Aa-20 in SSF and SMF with tannic acid and glucose as carbon sources. Pinto et al. (2001) investigated the tannase activity of 17 wild type and 13 mutant strains of *Aspergillus niger* and selected the potential tannase producers for maximum tannase production by solid state fermentation. Sabu et al. (2005) recorded an enzyme yield of 13.03 IU/g dry substrate (gds) in his research investigation involving tannase production under SSF using *A. niger* ATCC 16620 with palm kernel cake and tamarind seed powder as the substrate. Kumar et al. (2007) recorded maximum tannase production (30.2 U/ml) from *A. ruber* at 30.1 °C after 96 h of incubation with jamun leaves (*Syzygium cumini*) by SSF strategy. Manjit et al. (2008) reported maximum tannase production (174.32 U/g) using *Aspergillus fumigatus* MA with Jamun leaves as substrate at 25 °C, pH 5.0 and 96 h of incubation. Reddy and Kumar (2011) reported maximum tannase production (41.6 U/mg) from *A.terreus* using wheat bran as a substrate. Kulkarni et al. (2012) reported maximum activity (116 U/g dry substrate) of tannase from *A.oryzae* using mixed substrate (Jamun and Babul bark in the ratio of 4:6) through SSF. Nandini et al. (2014 (documented that food and agricultural residues such as corn husk, tamarind seed powder, banana peel, coconut coir, and spent tea powder are most suited natural substrates for both tannase and gallic acid production through SSF. Deepa et al. (2015) investigated production of tannase from *Aspergillus niger* using wood chips as substrate under SSF. Malgireddy and Nimma (2015) in their research investigation reported maximal tannase production from *Aspergillus terreus* with wheat bran as a substrate. Various natural tannin-containing substrates like wheat bran, coffee pulp and tea residue, tamarind seed powder, and rice bran have been efficiently utilized for maximal tannase production via SSF.

Statistical modeling of tannase production

The optimization of fermentation process is an important tool for the development of optimum parameters to scale-up the tannase production. The statistical modeling has been used for medium optimization and for understanding the relationship between different parameters with smallest number of experiments (Singh and Mukhopadhyay 2016).

There are several reports in literature emphasizing on optimization of tannase production using statistical modeling. Response surface methodology (RSM) and Taguchi methodology have been most widely used statistical tools for optimization of tannase production. Response surface methodology is utilized for determining the influence of factors over the

response and to optimize these variables to reach the highest productivity (Das et al. 2009). Taguchi methodology determines the influence of individual factors and reveals the correlation between the variables and process parameters (Taguchi 1986). Lekha and Lonsane (1994) optimized the variables such as initial pH, inoculum ratio, temperature, fermentation time, and moisture content using RSM for the production of tannase by solid state fermentation. Battestin and Macedo (2007) studied the optimization of variables (substrate concentration and % residue) through RSM. Sharma et al. (2007) proposed central composite rotatable design (CCRD) for optimization of tannase production from *Aspergillus niger.* Results revealed that tannic acid concentration, sodium nitrate, incubation period, and agitation rate were the most influencing factors for tannase production. Naidu et al. (2008) obtained twofold increase in activity of tannase from *Aspergillus foetidus* in SMF using a dual statistical approach involving a Plackett-Burman design to determine the key factors for production of tannase and RSM (utilizing a central composite design) to optimize these factors. Das et al. (2009) used taguchi method to optimize the concentration of tannic acid for tannase production from *Bacillus licheniformis* in SMF. Beniwal and Chhokar (2010) also optimized the parameters such as agitation rate, substrate concentration and incubation period for tannase production from *Aspergillus awamori* and *A. niger* in SMF through RSM. Mohan et al. 2014 used Plackett–Burman design and identified tannic acid, magnesium sulfate, ferrous sulfate and ammonium sulfate as significant nutrients affecting tannase production from *Aspergillus flavus* using tamarind seed powder as substrate in submerged fermentation. In addition, the optimization of process parameters using RSM resulted in maximum production of tannase (139.3 U/ml).

Xiao et al. (2015) optimized tannase production from *Aspergillus tubingensis* through sequential statistical approach under SSF.

Cloning and expression of tannase

Advancements in recombinant DNA techniques have widened the scope of creating genetically modified variants of existing microbial strains with desired enzyme machineries. Because of the several technical barriers involved in tannase production through traditional technologies, there has been a keen interest in molecular biology techniques for enhancing the production of tannase using recombinant microorganisms. Hatamoto et al. in Hatamoto et al. 1996 for the very first time clones and sequenced the gene encoding for tannase from *A.oryzae*. They further expressed this gene in *A. oryzae* strain having lower tannase-producing ability and reported as many as three fold increased production of tannase in transformants in comparison to wild strain. The increase in tannase production level in the transformant genome was due to additional gene encoding for tannase as confirmed by Southern blotting. Zhong et al. (2004) cloned and sequenced tannase-encoding gene from *Aspergillus oryzae* in the methylotrophic yeast *Pichia pastoris*. They reported significantly higher extracellular tannase production (7000 U/L) in the transformed yeast in SMF fed-batch production system utilizing glycerol and methanol as carbon source and transducer respectively. Cerda-Gomez et al. (2006) designed primers (Tan 1 and Tan 2) by using conserved sequences of tannase gene obtained from various species belonging to *Aspergillus.* They further used the primers set for amplifying a 435-bp DNA fragment taken from four distinct *Aspergillus* species through PCR. Noguchi et al. (2007) investigated the alliance of tannase-producing bacteria and colon cancer and explore novel gene encoding for tannase production. Iwamoto et al. (2008) identified the tannase-encoding gene from *Lactobacillus plantarum* from GenBank database on the basis of literature available about *L. plantarum* WCFS1. Tannase-encoding gene was then cloned and hyperexpressed in *Escherichia coli.* Recombinant tannase revealed a single protein of approximately 50.7 kDa after purification. On the other hand, Curiel et al. (2009) documented the production and purification of recombinant *Lactobacillus plantarum* expressed in *E. coli* using vector pURI3 and inserted the gene encoding for tannase with an aminoterminal His-tag. This strategy gave significantly higher amount of pure tannase (17 mg/L) by adopting single-step affinity method. Beena et al. (2010) isolated gene encoding for tannase from *A. awamori* and documented an ORF of 1122 bp upon sequencing. Homology studies conducted revealed a higher similarity index between *A. awamori* gene with that of *A. niger* in comparison to *A. oryzae* gene. Researchers are now emphasizing on metagenomic approach for identifying and exploring the tannase-encoding genes of microbes that are difficult to culture. Yao et al. (2013) documented a novel gene-encoding tannase (tan410) of 1563 bp from cotton field metagenomic library by functional screening. They cloned the tan410 gene and expressed it in *E.coli* BL21 (DE3) using pET-28a expression system under the control of T7 lac promoter. The recombinant tannase revealed interesting properties like 55 kDa molecular weight upon purification and characterization.

Tannins as substrates for tannase and gallic acid production

Tannins are high molecular weight (500 to 3000 kDa) polyphenolic compounds that exist abundantly in different parts o plants such as fruits, leaves, and bark (Aguilar et al. 2007; Rodriguez et al. 2008). Tannins are the seconds most copius polyphenols following lignins (Bhat et al. 1998). Aguilar et al. (2007) categorized tannins into four major groups:

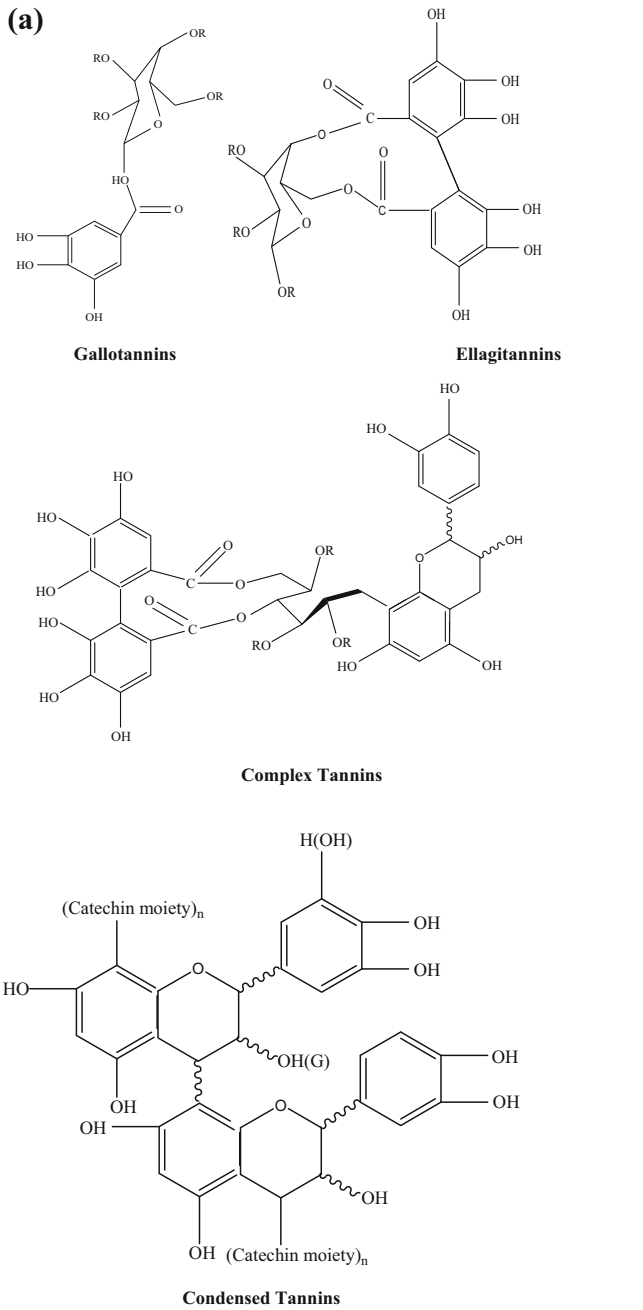

Fig. 2 a Classification of tannins. **b** Tannic acid structure

Table 3 Agricultural wastes as sources of tannin

Material	Tannin content (mg/g)	Reference
Rice bran	0.096	Paranthaman et al. (2009)
Redgram husk	2.601	Paranthaman et al. (2009)
Tea dust	0.102	Gowdhaman et al. (2012)
Blackgram husk	0.910	Arulnathan et al. (2013)
Black plum	38.37	Kumar et al. (2016)
Babul	41.6	Kumar et al. (2016)

gallotannins, ellagitannins, condensed tannins, and complex tannins. The structure of different tannins and tannic acid is depicted in (Fig. 2a, b.

Tannic acid is the commercial form of gallotannins. It mainly consists of glucose esters of gallic acid. Chinese gallotannin (*Rhus semilata)* is the principally utilized natural hydrolysable tannin for gallic acid production.

Tannins are present in several naturally occurring agricultural wastes such as redgram husk, greengram husk, blackgram husk, tamarind seed powder, tea dust, rice bran, and groundnut shell which can be utilized in industrial bioprocess for the production of value added products such as gallic acid through submerged fermentation (Table 3).

A number of research investigations utilizing a concoction of agro-industrial wastes like Paddy husk, wheat bran, Palm kernel cake, Cashew waste, apple baggasse, and rice bran for enhancing the tannase and gallic acid production have reported (Lekha and Lonsane 1994; Sabu et al. 2005; Battestin and Macedo 2007; Rodriguez et al. 2008; Paranthaman et al. 2010). Hydrolysable tannins are polyphenolic compounds consisting of gallic acid esters (gallotannins) or ellagic acid esters (ellagitannins) with a sugar core (generally glucose). Hydrolysable tannins can be easily hydrolysed under mild acid or alkaline conditions with hot water or enzyme (Lopez-Rios 1984). The principally utilized commercial sources of hydrolysable tannins include tannins from Chinese gall *(Rhus semialata),* Keekar (*Acacia nilotica*) leaves, Red gram husk and Cashew waste testa (*Anacardium occidentales*), and Myrobalan nuts (*Terminalia chebula*). Some of the plant sources that can be efficiently utilized for production of tannase and gallic acid have been listed in (Table 4).

Tannins as pollutants

Leather industry is one of the highest water-consuming industries generating extensively polluted effluent after processing of leather having high loads of pollutants (Song et al. 2004). Tannery waste water is reckoned as one of the highest ranked environmental pollutant amongst all forms of waste water generated from various industries (Verma et al. 2008; Gupta et al. 2012). Emergence of tannery waste water as an absolute pollutant in countries such as China has eventually posed catastrophic threat to mankind and aquatic life. Tannins are used in tanneries for processing of leather in the form of tannic acid. During tanning process significantly high levels of water, tannin

Table 4 Major plant sources for production of gallic acid

Source plant	Common name	Plant part	Type of tannin	Tannin %
Rhus coriaria	Sumac	Leaves	Gallotannin	10
Larrea tridentate	Creosote bush	Leaves	Ellagitannin	16
Caesalpinia cacalaco	Cascolate	Leaves	Ellagitannin	25
Anacardium occidentale	Cashew	Testa of seeds	Ellagitannin	10–40
Terminalia chebula	Myrobalan	Fruit	Ellagitannin	40
Caesalpinia spinosa	Tara Fruit	Pods	Gallotannin	40
Flurensia crenua	Tar bush	Leaves	Ellagitannin	40
Caesalpinia coriaria	Divi divi	Pods	Ellagitannin	43
Caesalipinia dignya	Teri pod	Pod cover	Gallotannin	45
Quercus infectoria	Turkish galls	Nuts	Gallotannin	81
Rhus semialata	Chinese galls	Galls	Gallotannin	89

*(Compiled from: Bajpai and Patil (2008); Banerjee et al. (2005); Deschamps and Lebeault (1984); Kar et al. (1999), Kar and Banerjee (2000) and Kar et al. (2002); Lokeswari and Jayaraju (2007); Paaver et al. (2010); Pourrat et al. (1987); Ruiz-Aguilar et al. (2004); Ventura et al. (2008), Lokeswari (2010), Lokeshwari (2016)

(in the form of tannic acid) and several other chemicals are used for processing of raw hides and skins. The entire process generates an approximate 30–35 m^3 volume of waste water per ton of raw hide/skins processed (Lofrano et al. 2008; Islam et al. 2014). In addition, the industries utilizing plant constituents as raw and processing materials eventually produce enormously high levels of waste water rich in tannins. The dark brown color of Tannery waste water causes blockage of sunlight thus reducing the photosynthesis and oxygenation activities of aquatic photosynthetic organisms and thus becomes fatal for them (Mwinyihija 2010; Carpenter et al. 2013). Consequently, the reduced dissolved oxygen level promotes the anaerobic conditions which results in unpleasant odor of aquatic organisms (Sahu et al. 2007; Verma et al. 2008). Tannery waste water also adversely affects the quality of water resources (Schilling et al. 2012; Dixit et al. 2015). Tannery waste water is rich in organic and inorganic components which may serve as nutrients for promoting the growth of several pathogenic bacteria thus severely contaminating the water resources (Verma et al. 2008; Bharagava et al. 2014). Common effluent treatment plant (CETP)-treated waste water has significantly high BOD, COD and TDS levels in addition to various other toxic pollutants like chromium which ultimately renders the water unsafe for mankind and other forms of life (Lofrano et al. 2013; Dixit et al. 2015). Thus, the currently available conventional chemical and biological waste water treatment methods are not good enough to remove these pollutants especially tannins because of their recalcitrant nature and low biodegrability. This necessitates the development and utilization of appropriate and effective treatment methods for bioprocessing of such enormously high volumes of tannery waste water. Bioremediation of these tannin rich waste waters using suitable enzymes may result in their effective biodegradation. The ability of microbial tannase to efficiently degrade natural tannins and tannic acid can be utilized for bioremediation of tannery waste water high in tannins and eventually for Gallic acid production.

Gallic acid production from tannins

Commercial production of gallic acid is undertaken by hydrolysing the tannins either chemically or enzymatically. The chemical approach for producing gallic acid generally consists of acidic hydrolysis of naturally occurring hydrolysable tannins (Mukherjee and Banerjee 2003). This particular approach is economically not feasible on account of reduced yield and lower purity of gallic acid (Bajpai and Patil 2008). In addition, this approach uses relatively higher concentrations of acid or alkali which causes corrosion of vessels utilized during the entire process. Thus, this approach requires better and effective safety means. One of the major drawbacks of this strategy is the generation of toxic effluents that are hazardous to environment thus ultimately posing threat to mankind (Banerjee et al. 2001).

As an alternate, the production of gallic acid is undertaken via microbial fermentation approach by cleaving the ester and depside bonds in hydrolysable tannins thus giving off gallic acid. The microbial tannase-based bioconversion of tannins to gallic acid offers several advantages over chemical technology in terms of ease of cultivation of cultivation of microorganisms, better control over various process parameters such as pH, temperature, degree of aeration as well as several environmental factors indispensable for the optimal growth of the microorganisms. The enzyme utilized mainly belongs to fungal or bacterial origin. The cell deprived fermentation broth is allowed to precipitate at reduced temperature so as to collect gallic acid. Gallic acid from fermentation broth is recovered via solvent extraction and is dried under vacuum till precipitation. This approach is economically feasible and environment friendly and does not cause any hazards to neither environment nor mankind.

Microbial biotransformation of tannins

Microbial degradation of tannins is most eminent approach for effectively biotransforming larger tannin molecules into relatively smaller molecules of higher market value. Tannin degradation potential significantly varies among different microbes like bacteria, fungi, and yeast. Yeast can effectively degrade gallotannins but looses its effectiveness in degrading elagitannins. Over the years, enzymatic hydrolysis of tannins by microbial tannases has gained momentum (Dhiman et al. 2017). Bacteria have stupendous potential to efficiently degrade gallotannins as well as ellagitannins (Deschamps et al. 1983). Fungi can effective depolymersise various tannins (Bhat et al. 1998). The enzymes involved in depolymerization of tannins include tannase and gallic acid decarboxylase. However, as a matter of fact, tannase has been the most widely investigated and utilized ezyme for tannin degradation. Microbial tannase holds utmost importance in tannin degradation as compared to tannase from plant and animal sources (Aguilar et al. 2007). Tannase breaks the ester and depside bonds in different types of tannins. However, their effectiveness in degrading condensed tannins is limited by their inability to affect C-C bonds (Haslam and Stangroom 1966). Gallic acid decarboxylase mediates the decarboxylation to gallic acid to pyrogallol; however, the enzyme is extremely unstable due to its relatively higher sensitivity to oxygen which renders its isolation and purification quite ardous (Zeida et al. 1998). There are certain bacteria like *Selenomonas gallolyticus* and *E.coli* that catalyzes decarboxylation of gallic acid to pyrogallol. Further transformation of this compound does not take

place possibily because of it having lesser toxicity or its production being thermodynamically more viable (Mingshu et al. 2006). In ellagitannin degradation, the liberation of ellagic acid is mediated by ellagitannin acyl hydrolase. However, extensive research investigations are required to evince the actual catalytic variability between tannin acyl hydrolase and ellagitannin acyl hydrolase and to comprehend the actual mechanism of tannin degradation (Aguilera-Carbo et al. 2008). The understanding of complex and condensed tannins is quite toilsome owing their convoluted structures. Thus, there has been only limited success in comprehending their degradation process. The degradation pathway of gallotannins is represented in Fig. 3.

Microbial tannase-catalyzed biotransformation of tannins can be efficiently utilized for producing significant levels of gallic acid. Gallic acid production and tannase production are interconnected with each other since tannase catalyzes the depolymerization of hydrolysable tannins thus releasing gallic acid.

Bacterial biotransformation of tannins

Some bacterial strains pertaining to genera such as *Bacillus* (Raghuwanshi et al. (2011), *Pseudomonas* (Selwal et al. 2010), *Staphylococcus* (Noguchi et al. (2007), *Klebsiella* (Sivashanmugam and Jayaraman 2013), *Lactobacillus* (Guzman-Lopez et al. (2009), *Citrobacter* (Wilson et al. (2009), *Serratia* (Belur et al. 2010), *Pantonea* (Pepi et al. 2010), Azobacter (Gauri et al. 2012), and *Enterobacter* (Mandal and Ghosh 2013) have been documented with the ability to degrade tannins. Lewis and Starkey (1969) documented the biodegradation of gallotannins from an aerobic bacterium *Achromobacter* sp. Deschamps et al. (1981) isolated several bacterial strains capable of degrading hydrolyzable and condensed tannins, including chestnut, wattle, and Quebracho commercial tannin extracts by enrichment. Bacteria tannase have the stupendous ability to hydrolyse natural tannins and tannic acid (Deschamps et al. (1983). Deschamps et al. (1983) documented the gallotannin (1% *w*/*v*) degradation by *Bacillus pumilus*, *B. polymyxa*, *Corynebacterium*, and *K. pneumonia.* Moreover, *B. pumilus*, *B. polymyxa*, and *K. planticola* utilized chestnut bark as substate and produced tannase followed by Gallic acid generation (Deschamps and Lebeault 1984). The anaerobic degradation of gallotannins mediated by a consortium of anaerobic sludge bacteria was first reported by Field and Lettinga (1987). Singh et al. 2001 reported gallic acid production through microbial degradation of tannic acid by ruminal fluid of cattle. Kachouri and Hamdi (2004) documented the potential of *Lactobacillus plantarum* in degradation of tannic acid. In most cases bacterial tannase utilizes methyl gallate as a substrate for producing gallic acid as the end product through oxidation process (Nishitani and Osawa 2003; Nishitani et al. 2004; Vaquero et al. 2004). Comprehensive tannin metabolism pathway is yet to be explored. Kumar et al. (1999) and Gauri et al. (2012) investigated the mechanism of tannic acid degradation and reported that glucose released by tannin degradation enters glycolysis and eventually TCA cycle. Gallic acid decarboxylase transforms gallic acid produced into pyrogalloll which is eventually converted to pyruvic acid, cis-aconitic acid, 3-hydroxy-5-oxo hexanoate and eventually enters the TCA cycle.

Fungal biotransformation of tannins

The role of fungi in tannin degradation dates back as early as 1900, when Fernbach Pottevin in 1900 independently reported the hydrolysis of tannins using cell free preparation of *Aspergillus niger* (Pottevin 1900). *Aspergillus*, *Rhizopus*, and *Penicillium* have been predominant filamentous fungi involved in the biotransformation of tannins. Along with them, various other fungi pertaining to genera *Trichoderma*, *Fusarium*, *Chaetomium*, *Rhizoctonia* etc. have also been reported for their ability to degrade the tannins especially the hydrolysable tannins. *Aspergillus* sp. has been the most potent and most widely studied tannase producer among other existing fungal sources. Fungal systems are well recognized for their capability to degrade hydrolysable tannins (gallotannins). Yamada et al. (1968) documented the evolution of tannin degrading systems in fungi belonging to genera *Aspergillus* and *Penicillium*. Ikeda et al. (1972) reported the interrelationship between tannin source and type of microbes being utilized for gallic acid production. Suseela and Nandy (1985) investigated the effect of various process parameters like pH, temperature, and cabon source on tannic acid degradation and Gallic acid production by *Penicillium chrysogenum*. The degradation of hydrolyzable tannins, specifically tannic acid has received a great deal of attention (Kumar et al. 1999). Kar and Banerjee (2000) documented proportionality in tannase and gallic acid production. Belmares et al. (2003); Cruz-Hernandez et al. (2009) reported the oxidative degradation of hydrolyzable tannins in *Aspergillus* sp. A good number of research studies involving fungal tannase have shown significant rise in tannin degradation in presence of carbon sources. It is a well established fact that tannase-catalyzed degradation of tannic acid gives off gallic acid and glucose. However, reserachers have succeeded in exploring pyrogallol as the intermediate compound of this metabolism along with gallic acid and glucose. Several research studies using various combinations of tannin containing substrates with major focus on optimizing the tannase and gallic acid production have been reported (Banerjee 2004). Several bacteria and fungi have been documented for biotransformation of tannins to Gallic acid (Table 5).

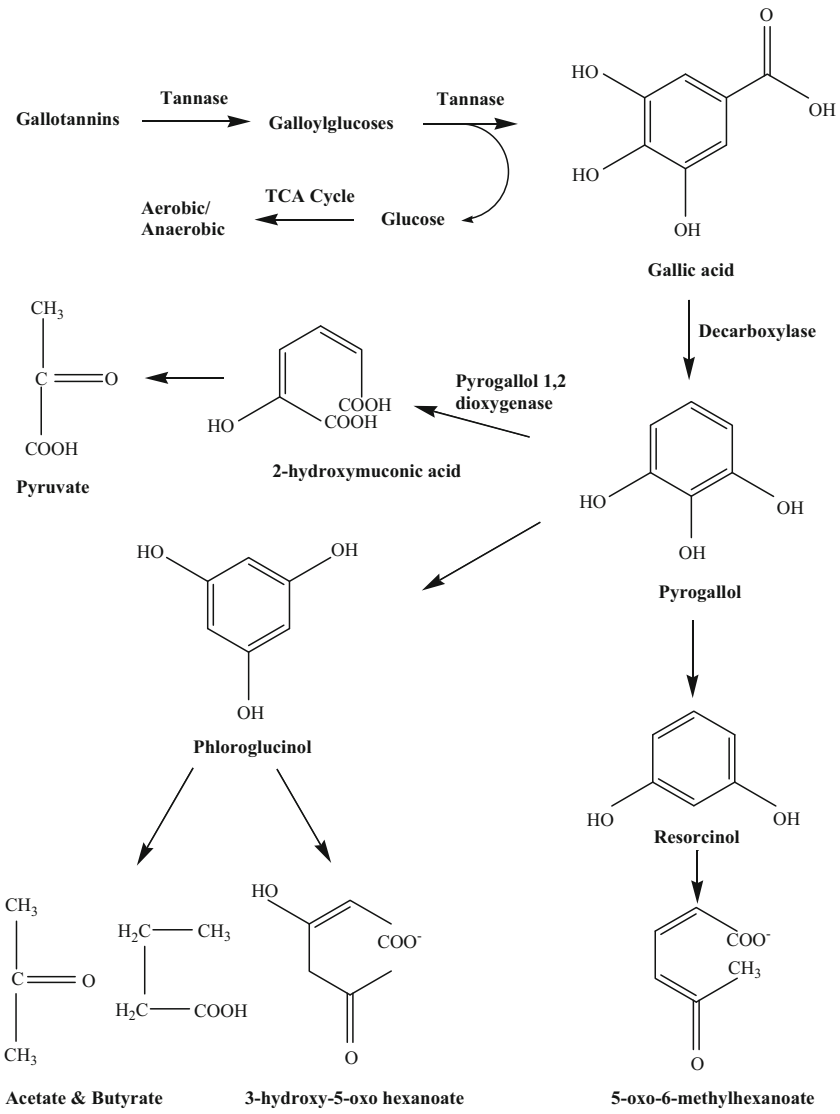

Fig. 3 Schematic representation of gallotannin biodegradation

Immobilization of tannase

Tannase can be immobilized by typical methods such as physical adsorption, covalent coupling, encapsulation, entrapment, or crosslinking. Immobilization facilitates tannase-enhanced catalytic activity and stability, reusability, easily controls the enzymatic reactions, product purification, and improved process economy (Ong and Annuar 2018). Tannase has been encapsulated in alginate, chitosan, carrageenan or pectin gel matrices.

Aspergillus oryzae tannase was immobilized by covalent binding of its glycosidic part on chitosan, chitin, Dowex 50 W, DEAE- sephadex A-25 (Abdel-Nabey et al. 1999). The highest enzyme activity was found on chitosan with a bifunctional agent (glutaraldehyde Immobilized tannase has been used for the gallic acid production (Mahendran et al. 2006; Sharma et al. 2008) and its esters in non-aqueous medium (Sharma and Gupta 2003; Yu et al. 2004). Sharma et al. (2002) non-covalently immobilized *A. niger* van tighem tannase on concanavalin A-Sepharose A-Sepharose via bioaffinity interaction. Sharma and Gupta (2003) successfully immobilized *Aspergillus niger* tannase on Celite-545 to produce propyl gallate. Yu et al. (2004) immobilized tannase on chitosan-alginate membrane by microencapsulation for the synthesis of propyl gallate. Das et al. (2007) utilized calcium alginate to immobilize *B. licheniformis* tannase. Chhokar et al. (2008) investigated immobilization of *Aspergillus heteromorphus* tannase on chitin, DEAE-sephadex A-50 and Ca-alginate. Among these supports, chitin was documented as best support matrix with 88% operational stability after seven cycles of reactions. Sharma et al. (2008) reported amberlite IR as best support for immobilizing the *P. variable* tannase. The immobilized enzyme retained catalytic activity even after six times reuse. Su et al. (2009) immobilized tannase on alginate by crosslinking-entrapment-crosslinking method and reported 86.9% residual activity after 30 times repeated use. Curiel et

Table 5 Gallic acid production by microorganisms

Microorganism	Substrate	Mode of fermentation	Incubation period (h)	Gallic acid yield	Reference
Aspergillus niger	Tara fruit pods	Submerged fermentation	45	30[a]	Pourrat et al. (1985)
Aspergillus niger	Sumac leaves	Submerged fermentation	40	9.75[a]	Pourrat et al. (1987)
Aspergillus niger	Gall nuts	Submerged fermentation	24	40.5[a]	Regerat et al. (1989)
Rhizopus oryzae (free cells)	Tannic acid	Submerged fermentation	96	83.5[a]	Misro et al. (1997)
Rhizopus oryzae (immobilized cells)	Tannic acid	Submerged fermentation	96	78.5[a]	Misro et al. (1997)
Rhizopus oryzae	Powder of teri pod cover	Modified Solid state fermentation	72	90.9[a]	Kar et al. (1999)
Aspergillus awamori	Tannic acid	Submerged fermentation	30	40.3[c] (88%)[a]	Seth and Chand (2000)
Rhizopus oryzae	Myrobalan and teri pod cover (mixed substrates)	solid-state fermentation	60	85.67[a]	Mukherjee and Banerjee (2004)
Aspergillus foetidus	Myrobalan and teri pod cover (mixed substrates)	solid-state fermentation	72	90.48[a]	Mukherjee and Banerjee (2004)
Rhizopus oryzae and *Aspergillus foetidus*	Mixed substrate(Myrobalan and teri pod	Modified solid-state fermentation	48	94.8[a]	Banerjee et al. (2005)
Aspergillus aculeatus	*Cassia siamea*	Submerged fermentation	36	56.6[a]	Banerjee et al. (2007)
Aspergillus niger Aa-20	*Larrea tridentata*	Solid-state culture	48	0.4[c]	Trevino-Cueto et al. (2007)
Aspergillus fischeri	Tannic acid	Submerged fermentation	37	0.56[c]	Bajpai and Patil (2008)
Aspergillus niger 3	*Quercus infectoria*(oak)	Submerged fermentation	48	91.3[a]	Sarıozlu and Kıvanc (2009)
Penicillium spinulosum	*Quercus infectoria(*oak)	Submerged fermentation	48	93.2[a]	Sarıozlu and Kıvanc (2009)
Phomopsis sp. SX10	*Acer ginnala*	Submerged fermentation	168	200.47[b]	Qi et al. (2009)
Enterobacter cloacae MTCC 9125	Tannic acid	Submerged fermentation	48	3.47[d]	Beniwal et al. (2010)
Enterococcus faecalis	Tannic acid	Submerged fermentation	24	0.28[d]	Goel et al. (2011)
Bacillus sphaericus	Tannic acid	Submerged fermentation	24	90.8[a]	Raghuwanshi et al. 2011
A. niger AUMC 4301 extracellular tannase	Mango seed kernel	Solid State Fermentation	18	103.6[e]	El-Fouly et al. (2012)
Bacillus subtilis PAB2	Tannic acid	Submerged fermentation	36	6.45[c]	Jana et al. (2013)
Lactobacillus plantarum CIR1	Tannic acid	Submerged fermentation	24	8.63[c]	Aguilar-Zarate et al. (2014)
Bacillus subtilis AM1	Tannic acid	Submerged fermentation	12	2.416[c]	Zarate et al. (2015)
Penicillium crustosum AN_3 KJ820682	*Pinus*(Pine needles)	Solid State Fermentation	120	9.29[e]	Thakur and Nath (2017b)

[a] %, [b] mg/liter, [c] g/liter, [d] mg/ml, [e] mg/g

al. (2010) covalently immobilized recombinant *L. plantarum* tannase onto glyoxyl agarose and the enzyme was able to retain its catalytic activity after 1 month. *Aspergillus niger* GH1 tannase has been immobilized by entrapment in calcium alginate beads (Flores-Maltos et al. 2011). Tanash et al. (2011) immobilized the tannase from *Aspergillus aculeatus* on gelatin by cross-linking and observed the highest activity. Yao et al. (2014) immobilized Tan410 tannase of metagenomic isolate on silica SBA-15, calcium alginate, chitosan, and amberlite IRC 50. Results revealed calcium alginate as the most suitable support and immobilized enzyme retained its activity after 1 month. Kumar et al. (2015) studied the immobilization of *Aspergillus awamori* tannase on various supports such as Dowex 50, pectin, sephadex 25–40, DEAE–sephadex, amberlite, silica, and chitin. Among them, amberlite proved as best support for immobilization. Wu et al. (2016) immobilized *Aspergillus tubingensis* tannase onto carboxyl-functionalized Fe_3O_4 nanoparticles (CMNPs). Lima et al. (2017) covalently immobilized tannase onto magnetic nanoparticles composited with polyaniline coated diatomaceous earth. Li et al. (2018) immobilized *Aspergillus niger* tannase by a glutaraldehyde conjugate with chitosan-coated magnetic nanoparticles.

Scientific perspectives

Deschamps et al. (1983) reported that the fungal and yeast tannase exhibit considerably good activity in biodegrading the hydrolysable tannins, however lesser activity in the degradation of natural tannins. The drawback of utilizing fungal strains in industrial applications is that degradation by fungi is slow and they are not easy to genetically alter because of their genetic complexity (Beniwal et al. 2010, 2013). It has been documented that bacterial tannase can effectively hydrolyse natural tannins and tannic acid. Bacteria can efficiently degrade gallotannins as well as ellagitannins. Furthermore, bacteria possess the capability to live under extreme temperature conditions, thus they may prove as a potential source of thermostable tannase (Beniwal et al. 2013). Although several reports regarding fungal and bacterial tannase have been studied and it is evident that fungal tannase has been extensively studied and exploited for tannase and gallic acid production in comparison to bacterial tannase. As a matter of fact, in the last 150 years of tannase research (1867–2017), most of the researchers have drawn a straight forward portrait on tannase from fungi rather than bacteria. However, the bacterial tannase on the other hand personifies a stupendous potential for industrial utilization. More than five billion metric tons agrowaste biomass is being generated per year in the form of fruits and vegetable wastes, wheat bran, sugarcane bagasse, cotton leaf scraps, rice bran, rice straw, ground nut cake, cashew waste, tamarind seed, pomegranate rind etc. Currently in India, more than 960 million tons of solid waste is being produced annually as byproducts during industrial, mining, municipal, agricultural and other processes. In India, more than 40% of solid waste generated annually is from organic and agricultural sources. There is a growing concern for these accumulating wastes as they are either being dumped in landfills, burnt, or left to rot in the open, leading to severe environmental pollution. A sustainable solution would be to utilize these solid wastes as an "economical" alternative to costly raw materials to effectively and economically produce industrially important products of industrial and commercial potential such as gallic acid (Nandini et al. 2013; Wang et al. 2016).

Conclusions

The industrial importance of gallic acid is well established. There are two possible routes of for gallic acid production at industrial level: chemical technology and enzyme-based technology. From our review, we concluded that enzyme-based technology has a clear cut upper edge over chemical technology of gallic acid production. The bioconversion by enzyme as well as whole cell biocatalyst has tremendous importance in industry owing to escalated yields, low impurity profiles, environmental safety, and process reproducibility.

The original studies on fermenter scale biotransformation of tannins to gallic acid using microbial tannase are scanty. This is limited by several constraints like limitation in using high tannin concentration due to to sensitiveness of the microbes (being used) to tannic acid, dearth of information on exact tannin metabolism and properties of tannase etc. The superfluity of tannin-containing agro- and industrial waste demands more efficient tannase that would be able to withstand higher concentration of tannic acid at fermenter scale. Thus, more research is needed in areas concerning studies to lessen the sensitivity of microorganisms to tannic acid, to understand the complete tannin metabolism, to enhance the hydrolysis rate of tannins, to establish a superior process control, and to develop economically feasible, environment friendly, more productive and proficient biotransformation processes for production of gallic acid for industrial and commercial utility.

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