

Recent developments in bio-based adhesives from renewable natural resources

Ana Arias^{1,*}, Sandra González-Rodríguez¹, Murillo Vetroni Barros², Rodrigo Salvador², Antonio Carlos de Francisco², Cassiano Moro Piekarski², María Teresa Moreira¹

¹CRETUS Institute, Department of Chemical Engineering, Universidade de Santiago de Compostela, Spain.

²Sustainable Production Systems Laboratory (LESP), Graduate Program in Industrial Engineering (PPGEP), Universidade Tecnológica Federal do Paraná (UTFPR), Ponta Grossa, Paraná, Brazil

*Corresponding author

E-mail address: anaarias.calvo@usc.es

Abstract

Wood-based panels play an important role in the furniture and decoration industries worldwide. However, the adhesives used in the manufacture of those panels are a subject of concern. Therefore, the aim of this study is twofold: (i) to critically analyze the available reports on the formulation of bio-based adhesives for the manufacture of wood panels, evaluating the different renewable raw material options, and (ii) to identify the advantages, disadvantages and difficulties encountered that hinder further development. For this purpose, the *Methodi Ordinatio* was used to assist identifying high impact research on the topic, and the VOSviewer software was used for analyzing the co-occurrence of keywords. Visual graphs were used to discuss the main research themes, type of study, publications per year, and publications per journal. This served to draw on the main aspects of the referenced body of literature. This review provides a rationale for showing the suitability of the methodological approaches and research designs used, as well as the production of bio-adhesives under the premises of synthesis routes, techniques and procedures. In view of the potential development of bio-adhesives with the prospect of market implementation, two approaches are essential to ensure a step forward in their development at full scale, namely techno-economic evaluation and the assessment of the environmental impacts associated with the production of bio-adhesives based on the life cycle assessment methodology.

Keywords: wood-based panels; bio-based adhesives; wood adhesives; renewable raw materials; review

1. Introduction

In recent years, organizations have been increasingly seeking to use materials and energy from renewable sources. Among the aims of such organizations, one can mention the optimization of processes and minimization of environmental impacts to be in their strategic core, so that competitiveness and sustainability are targeted with equal importance (França et al., 2021). In

39 that sense, wood has been a biomaterial of special interest due to its reported low impacts
40 related to climate change (Head et al., 2020). Wood is a renewable natural resource with a
41 complex polymeric structure consisting of lignin, cellulose and hemicellulose that is and has
42 been widely used for the production of a vast range of products (Kirk-Othmer, 1998). One of the
43 sectors in which its use is most widespread is in the field of wood panels such as plywood,
44 fiberboard and particleboard, among others (Zhang et al., 2019). On the one hand, a benefit
45 related to the use of wood as a raw material is attributed to its role as a carbon sink in the
46 development of the tree population (Ruter et al., 2008). Thus, it is established that, for each
47 cubic meter of wood, approximately 1 ton of CO₂ is stored. On the other hand, the use of wood
48 as a raw material for manufacturing activities leads to a reduction in the use of non-renewable
49 fossil resources, thus avoiding the environmental impacts associated with their extraction,
50 transport and processing activities.

51 A number of drawbacks associated with the manufacture of panel products have also been
52 listed, mainly due to the use of certain chemicals such as synthetic-based adhesives required in
53 the gluing phase of the boards. The most extensively adhesive used in plywood and
54 particleboard is urea-formaldehyde (UF) (Pocius, 2012), and in the case of fiberboard products,
55 phenol-formaldehyde (PF) and melamine-urea-formaldehyde (MUF) adhesives are the most
56 widespread (Mathias et al., 2016). More than half of the total world production of formaldehyde
57 is consumed for the manufacture of UF, MUF, and PF adhesives used in wood-based panels.

58 The high functionality of adhesives in relation to their excellent bonding properties and board
59 quality and strength gives them a dominant position, which greatly hinders the introduction and
60 development of natural-based, renewable, adhesives that do not include formaldehyde in their
61 formulation. Nonetheless, the fact that adhesive manufacturing processes are fully optimized
62 based on the operational conditions of pressure, temperature, and curing time undoubtedly
63 gives them an advantage over new developments. However, the research and development of

64 highly functional adhesives capable of replacing those based on formaldehyde has become a
65 necessity given the obligation to adapt to regulations limiting formaldehyde and volatile organic
66 compound (VOC) emissions at different stages of the life cycle of wood-based panels (Salem and
67 Böhm, 2013). In particular, the International Agency for Research on Cancer classified
68 formaldehyde as a carcinogenic substance for humans (Popović et al., 2020).

69 Investigating in more detail the stage associated with the manufacturing process, the main
70 direct and indirect emissions have been identified in a life cycle assessment study of
71 petrochemical adhesives (Wilson, 2010). Thus, the amount of formaldehyde released as an
72 emission derived from the manufacturing process is established in values around $7 \cdot 10^{-3}$ mg/kg
73 adhesive for emissions to air, and up to 10 times higher: $78.4 \cdot 10^{-3}$ mg/kg for emissions to water.
74 However, in addition to direct emissions, greenhouse gases (GHGs) associated with energy and
75 fuel requirements stand out, as well as releases of other VOCs, methanol, and phenol, which are
76 also considered hazardous air pollutants (HAPs).

77 The negative effects derived from the use of formaldehyde as a bonding agent in the wood panel
78 industry, the obligations derived from environmental and health regulations, as well as the need
79 for innovation, development and research of new natural products with the target of reducing
80 the consumption of fossil resources, are the main factors driving the substitution of
81 formaldehyde-based adhesives with bio-adhesives.

82 Moreover, there are numerous studies that propose the development of new formulations of
83 bio-based adhesives for use in the gluing phase of panel manufacturing, such as those based on
84 lignocellulosic biomass (including Kraft lignin and Organosolv) (Antov et al., 2021; Aziz et al.,
85 2019; Chen et al., 2020; Gadhave et al., 2019a), tannins (condensed and hydrolyzed) (Chen et
86 al., 2020; Tomak and Gonultas, 2018; Zhou and Du, 2020), soybean (soybean, soybean meal, and
87 soy protein) (Chen et al., 2017; Eslah et al., 2016; Li et al., 2017) and starch (corn) (Chen et al.,
88 2019; Gu et al., 2019; Li et al., 2020; Sun et al., 2018). From the point of view of commercial use,

89 tannin bio-adhesives have reached a better position in the market, as some formulations are
90 being developed in large-scale industrial manufacturing (Valenzuela et al., 2012; Zhou and Du,
91 2020).

92 Among the extensive literature consulted, a common trait is observed among the diversity of
93 research articles, the research has been developed on a laboratory scale, with a focus on the
94 chemistry and reaction strategies in the formulation of bio-adhesives as well as the
95 characterization of the developed products, and the study of the mechanical properties of the
96 boards. In this context, no significant number of references were found that address the
97 production process of bio-adhesives at pilot or industrial scale, so that techno-economic studies
98 have been used to extrapolate the laboratory results to a demonstrative or real process scale
99 (Arias et al., 2021).

100 The aim of this manuscript is to conduct a critical and extensive analysis of the work carried out
101 in the framework of bio-adhesives production from two complementary perspectives. On the
102 one hand, the review of the most recent developments in the manufacture of bio-adhesives, not
103 only from the perspective of the use of renewable raw materials, but also as valuable
104 information to identify prospects for large-scale development. Moreover, the environmental
105 valuation of bio-adhesives as a counterpart to synthetic resins is an additional reason supporting
106 the development of this type of bio-products.

107 Accordingly, this review is based on an analysis of the research studies developed within this
108 topic, using software services such as Mendeley Desktop®, VosViewer®, and Microsoft Power
109 BI®, which allowed the analysis of the type of publications on the topic and the visualization of
110 the most relevant data, providing rigorous information on the current development of the
111 alternative based on sustainable, environmentally friendly, and high-quality bio-based
112 adhesives.

113

2. Wood adhesives from renewable resources and free of formaldehyde

In order to provide adhesives suitable for the manufacture of good quality wood-based panel products it is essential to take into account their degree of penetration into the porous network of wood (Ülker, 2016), since both the quality of bonding and the formation of adhesive-wood intermolecular bonds directly influence on the mechanical properties of the wood-based panel, i.e. strength, water resistance, failure, which, on the other hand, also has to meet the requirements of the standards. Strength appears as one of the mechanical properties on which the different experimental studies for the development of new sustainable bio-adhesive alternatives are focused, not only because of the number of standards and specifications referring to this property (i.e. ASTM D905-08(2021), D906-20, D2339-20 GB/T 33333-2016), but also because it is one of the mechanical properties with the highest contribution on the final quality of the wood panel. Along with the bending properties and the modulus of elasticity, strength is the mechanical property in which the greatest variations are observed depending on the environmental conditions to which the wood board is exposed (Ayrilmis et al., 2010). Another crucial property that determines the applicability of wood panels, i.e., interior or exterior use, is moisture tolerance. Many experimental studies focus on the development of adhesives that are capable of developing wood-adhesive intermolecular bonds strong enough to reduce fluctuations in board size caused by moisture absorption from the environment. In many of these studies, crosslinking or hardening agents are used, providing a wood product with increased resistance to wet conditions.

Therefore, the development of new sustainable and bio-based adhesives should not only focus on reducing fossil resources and the impacts associated with their production compared to synthetic resins, but their study should focus on providing an adhesive that, in addition, complies with all standards, thus giving wood products with adequate technical and mechanical

138 properties. On the other hand, the production costs of bio-adhesives should be analogous or
139 similar to those of synthetic base, and also with production schemes.

140 2.1 Main classification of bio-adhesives according to the renewable material used

141 According to the criteria just mentioned, the different bio-adhesives alternatives for wood
142 manufacturing could be classified according to the renewable raw material used in their
143 formulation. Five main groups could be identified (**Figure 1**): tannin, lignin, wood, plant, soy and
144 starch adhesives and bio-adhesives. To clarify the concepts that will be used throughout this
145 review, when referring to adhesives, without the term "bio" it means that chemicals are used in
146 the formulation of the adhesive, for example for those adhesives where lignin can partially or
147 fully replace phenol in phenolic resins, but formaldehyde is still present in the adhesive
148 formulation.



149

150 Figure 1. Bio-alternatives for the production of wood bio-adhesives.

151

152 3. Methodology

153 The methodological procedures used in this piece of research were conducted according to a
154 number of steps. First, the relevant existing literature on bio-based adhesives used for the

155 manufacture of wood-based panels were searched and selected to comprise the research
 156 portfolio for this literature review (see section 2.1). In addition, the procedures followed to
 157 create visual maps can be seen in section 2.2.

158 **3.1 Methodi Ordinatio**

159 This section details the methodological procedure followed for collecting scientific publications
 160 that address the topic of bio-adhesives from different targets and methodologies. From a
 161 preliminary selection, it is necessary to conduct a critical analysis of the contributions. The
 162 methodology for systematic reviews called *Methodi Ordinatio* (Pagani et al., 2015) was applied,
 163 which allows determining the relevance of the documents. The main steps followed to perform
 164 the literature review are depicted in Table 1.

165 Table 1. Steps to conduct the literature review.

Database search	I	Database	Scopus
		Keywords	>100
		Number of documents	321
Filtering procedures	II	Elimination of duplicates and documents written in a language other than English	
	III	Screening title and keywords.	
	IV	Reading of abstracts.	
	V	Reading of full-texts.	
		Final Portfolio 275 documents 31 keywords	
Content analysis	VI	Type of article	
		Keywords	
		Year of publication	
		Journal	

166

167 **Step I. Database search.** A search was conducted in the Scopus database in January 2021,
168 using the sets of keywords presented in Figure 1, so that all research and review articles
169 published from the year 2000 to the date on which the search was performed were selected.

170 **Step II. Reference management.** All entries were managed using Mendeley, where all
171 duplicates and documents not written in English were removed.

172 **Step III. Screening title and keywords.** All the titles and keywords of each document were
173 read to allow selecting the articles according to their suitability to the research topic.

174 **Step IV. Reading of abstracts.** As with the titles and keywords, all the abstracts were read
175 to allow selecting the articles according to their suitability to the research topic.

176 **Step V. Reading of full-texts.** In this step, the full-texts of the articles selected were
177 retrieved and read. The decision for keeping or ruling out the full-texts was also based on
178 the premise of Step IV.

179 **Step VI. Content analysis.** The *Methodi Ordinatio* was used to calculate the *InOrdinatio*
180 coefficient that allows weighting the relevance of the documents taking into account their
181 number of citations, year of publication, and impact factor. The authors used the number
182 of citations of each article (found through Google Scholar on February 08, 2021) and the
183 Impact Factor (IF) (JCR) (2019) of each journal to determine the most relevant, high impact
184 research. The *InOrdinatio* coefficient can be seen in Table A.1 (Appendix A).

185 The manuscripts found (275 documents) were analyzed according to: (i) year of publication;
186 (ii) journal of publication; (iii) area of application; (iv) category of study; (v) Impact Factor;
187 and (vi) number of citations. The characteristics of the articles are detailed in **Table 1SM**.

188 **3.2 Visual graphics**

189 This section describes the procedure used to create visual maps using the VOSviewer v1.16.15
190 software tool. The settings used for the keyword co-occurrence map (Figure 2) are as follows:

- 191 - Map based on bibliographic data;
- 192 - Type of analysis: co-occurrence;
- 193 - Unit of analysis: keywords;
- 194 - Counting method: full counting;
- 195 - Minimum number of occurrences of a term: 5;
- 196 - Number of terms selected: total number of items;
- 197 - Show: all items.

198

199 A selection of 275 articles were compiled after the procedure described above. Moreover, the
200 keywords in the articles from the final portfolio were standardized according to the topics
201 developed in the manuscript so that it was possible to detect duplications derived from the
202 wording of the keywords (see further details in section 3.2). In addition, Flourish (**Figures 1 and**
203 **3**) and Microsoft Power Business Intelligence (BI) desktop (**Figure 4**) were used to build the
204 graphs and allow a clearer interpretation.

205 **4. Synthesizing the characteristics of the existing literature (final portfolio of documents)**

206 **4.1 Type of article**

207 An analysis of the topic and typology of the publications was the first stage in the review of the
208 bibliography. Eight categories have been identified according to the type of study developed.
209 The first category, and the one that reflects the main researchers interesting on the topic, is
210 based on experimental studies, i.e., those involving new adhesive formulations from renewable
211 raw materials in laboratory-scale tests. The main objective of these research works is to verify
212 how it is possible to activate the starting raw material through not only chemical but also
213 enzymatic stages in order to produce bio-adhesives with compatible characteristics to those of
214 synthetic resins used for the manufacture of boards (**Figure 2**).

215 A significant number of reviews publications have also been identified. Those studies address
216 the synthesis of bio-adhesives but also the follow-up of ongoing research that arouses the
217 greatest interest and where significant progress on the technique is envisioned. These
218 publications an overview and a broad approach, so that they not only represent the gathering
219 of manuscripts on the topic, but also show the objectives, scope, and rationale underlying
220 different research works that represent relevant progress. Special mention should be made to
221 the group of publications that aim to present future perspectives, which includes those
222 references that develop future perspectives and research options for the evaluation of bio-
223 adhesive production.

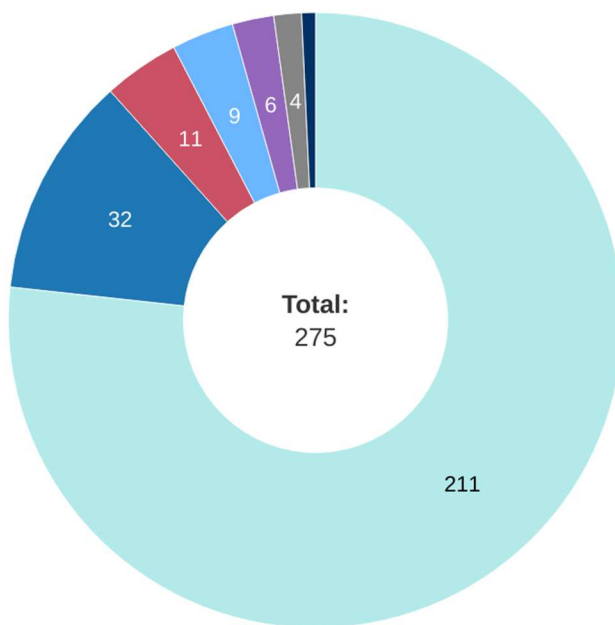
224 One research category of interest corresponds to the group of patents that have been developed
225 in the field of bio-adhesives. Although, to date, the development of patents is not extensive, it
226 is important to take them into account as they provide the overview of the prospect of bio-
227 adhesives in the context of industrial development.

228 In view of the potential development of bio-adhesives with the prospect of market
229 implementation, two approaches are essential to ensure a step forward in their development at
230 full scale. Techno-economic evaluation is the stage of conceptual design of the process under
231 development as well as a requirement in the proof of concept that allows the selection among
232 the most suitable alternatives according to technological and economic feasibility. It is also
233 relevant the identification of the environmental impacts associated with the production of bio-
234 adhesives based on the life cycle assessment (LCA) methodology.

235 The pie chart in **Figure 2** represents the number of articles available in the portfolio in each of
236 the categories mentioned above. As can be seen, the category of "experimental" studies stands
237 out, in which a total of 211 articles have been selected. The exhaustive analysis in the
238 subcategories shows several alternative raw materials for the formulation of bio-adhesives. The
239 most prominent are soy and lignin, with 66 and 49 publications, respectively, followed by tannin

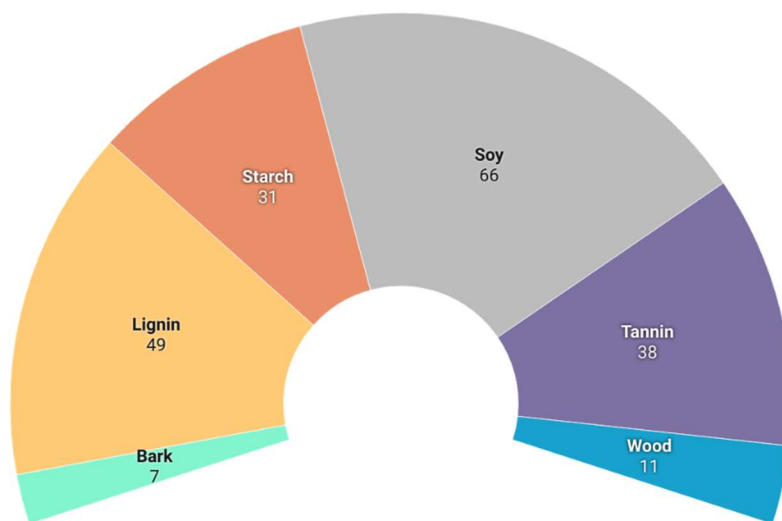
240 and starch. However, there are a large number of manuscripts reflecting the possibility of
241 combining two natural resources such as those based on tannins and soy. After the category of
242 experimental studies, the category of reviews, in terms of prospective developments, is the one
243 with the largest number of studies available, with a total of 32 studies out of the total analyzed.
244 Moreover, it is noteworthy that techno-economic evaluations and LCA studies are those in which
245 there are fewer references available. The reason for the low availability of this type of reports
246 lies in the scarce development of bio-adhesive production on a real process scale. Most of the
247 works carried out focus on experimental development at laboratory scale, which makes it
248 difficult to carry out a techno-economic or environmental analysis, given the lack of relevant
249 information, such as the sizing of equipment or energy requirements, data necessary for the
250 performance of such evaluations.

Experimental Review Book chapter LCA Book Patent TEA



Number of publications

251



252

Figure 2. Circular diagrams representing the number of each typology of publication and the main renewable raw materials identified for the experimental studies considered for the literature analysis. Created with: DataWrapper

253

254

255

256

257

258

259 **4.2 Keywords**

260 In a first attempt to show graphically the occurrences among the keywords of the manuscripts
261 selected, given the lack of standardization in terms of the selection of keywords in the writing
262 of the manuscripts, it resulted in a large number of words, which made the graphic difficult to
263 read, due to variability of terms that can be used to refer to the same concept. To illustrate this
264 fact, the following example is included: in the case of soy-based adhesives, the following
265 variability was found in terms of the keywords: soybean, soy flour, soy protein isolate, soy
266 protein concentrate, defatted soy flour, soy protein, soy flour-based adhesive, among others. In
267 accordance with the main objective of this review, which is based on a detailed analysis of the
268 available literature to select the types of bio-adhesives on which the research studies have
269 focused, this grouping of keywords refers to the same concept, i.e., soy bio-adhesives, since soy
270 is the raw material used for their formulation. Thus, the standard keyword selected to refer to
271 these publication keywords is soybean bio-adhesives. After grouping the different keywords, a
272 more suitable and understandable graphic visualization has been achieved, since the variability
273 of keywords used to refer to the same article has been significantly reduced. All of them are
274 represented in **Figure 3**.

275 **4.2.1 Raw material with the highest relevance according to keyword co-occurrence**

276 The results reported show that, among all the raw materials for the formulation of bio-
277 adhesives, soybean is the one that stands out. Equally noteworthy are those developed from
278 lignin, tannins and starch, showing a less pronounced development those formulated from bark
279 and wood fibers. However, two keywords: wood adhesives and wood bio-adhesives, have been
280 identified. The first term includes those adhesives that combine bio-based materials and
281 synthetic products such as urea, phenol, or formaldehyde. This code word is used in
282 experimental studies in which it has been proposed to partially substitute these chemicals with
283 lignocellulosic, protein or polymeric sources. However, given the percentage content of

284 synthetic components, these wood adhesives cannot be characterized as bio-adhesives. The
285 second term encompasses those experimental studies in which, unlike the previous ones, no
286 synthetic adhesive is included in the formulation; accordingly, phenol, urea, melamine, or
287 formaldehyde are included in their composition.

288 4.2.2 **Quality criteria of bio-adhesives and wood-boards according to keyword occurrence.**

289 Notwithstanding, most of the experimental studies include an analysis of the thermo-
290 gravimetric properties of the boards manufactured with the different bio-adhesive
291 formulations, which is one of the common sections that can be identified in all the bio-adhesives
292 experimental studies. These analyses make it possible to identify not only the percentage
293 compositions of the raw materials to be used for the development of a higher quality bio-
294 adhesive, but are also very useful in determining whether the development of wood panels
295 glued with these bio-adhesives complies with the standards and specifications of wood
296 products. The most relevant properties that have a direct effect on the final quality of the wood
297 board are those related to the bond strength between the adhesive and the wood fibers,
298 resistance to water, since this parameter will determine the final use of the board (indoor or
299 outdoor use), modulus of rupture, being a property of high importance for those wooden boards
300 whose purpose is its use in the field of construction, and shear strength, both in dry and wet
301 conditions.

302 4.2.3 **Chemicals with the highest occurrence scores in the keywords**

303 Most adhesives require the addition of so-called "crosslinking agents", which are compounds,
304 usually chemical-based (but can also be bio-based), that develop crosslinking bonds between
305 the raw materials used for their formulation. The most widespread is glyoxal, since its use allows
306 increasing the dispersion and solubility of the polymeric structure of lignocellulosic and protein
307 materials without requiring chemical modification (Park et al., 2017). On the other hand, its use
308 as a crosslinking agent, in addition to providing an improvement in the mechanical properties of

325 interactions among the most recurrent keywords: "water resistance", "strength", "mechanical
326 properties". As for the type of board used for the development of the experimental studies, the
327 most relevant are plywood and composites, followed by particleboards. On the other hand, it is
328 equally important to consider how "environmentally friendly" the adhesives may be, both from
329 the point of view of the chemicals required in their formulation, and from the perspective of
330 electricity and heat consumption.

331

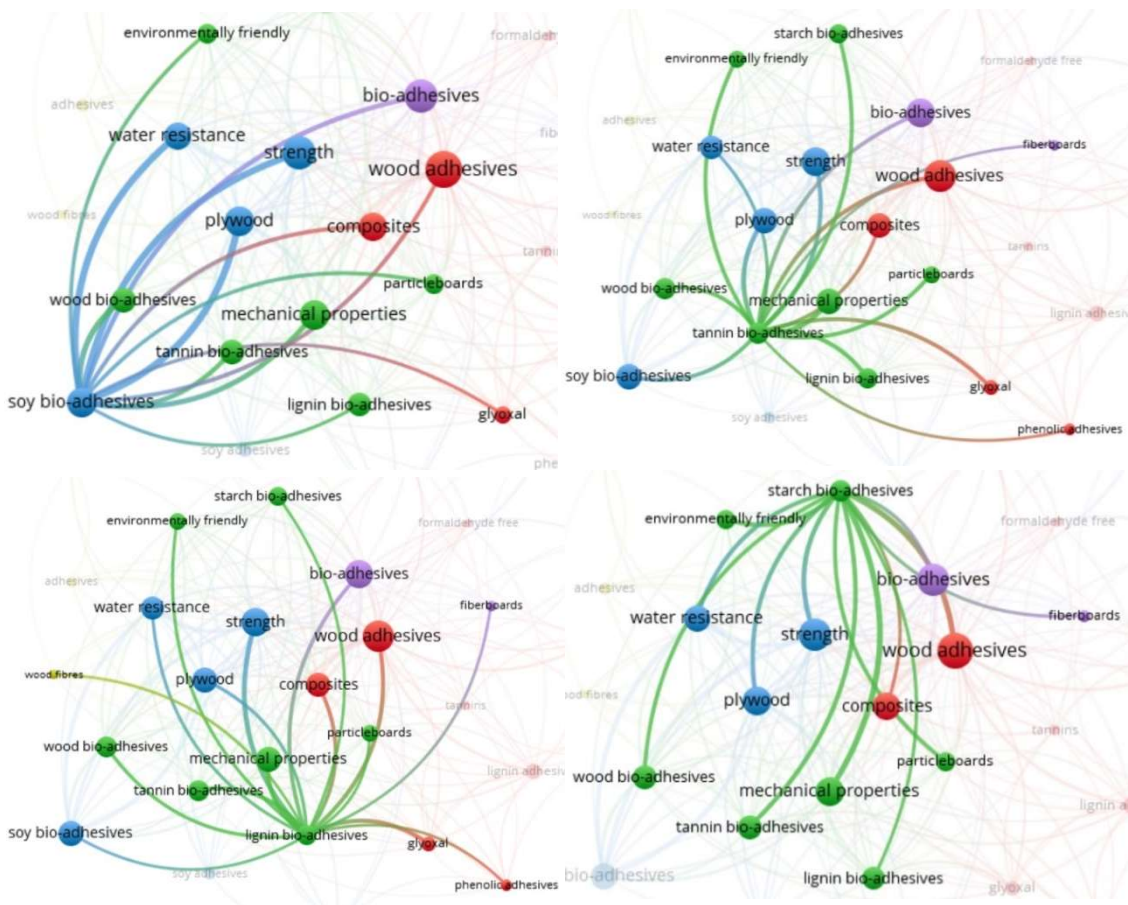


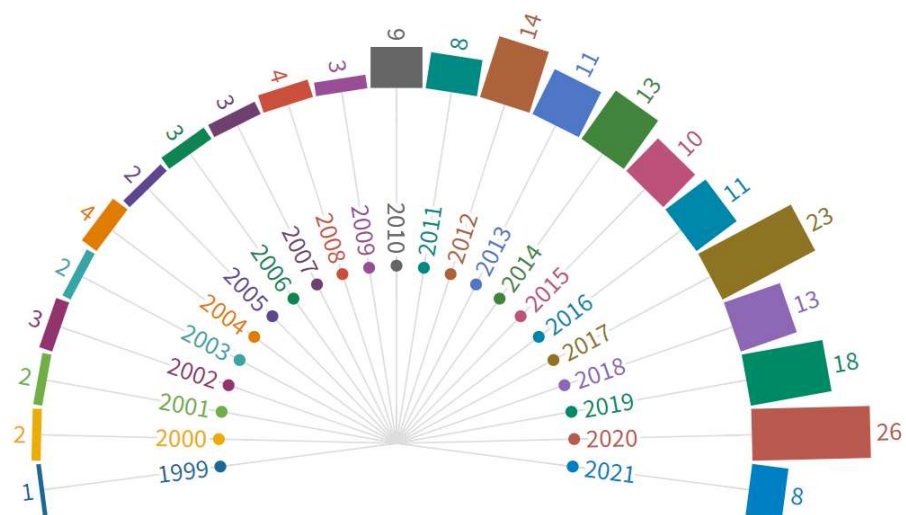
Figure 4. Co-occurrence graphical abstracts regarding the main raw materials used for the development of bio-adhesives.

332

333 4.3 Year of publication

334 The first report describing the development of new natural-based materials that could be
335 applied in the wood-based panel production process was published in 1999 under the title

336 "Commercial use of lignin-based materials" and was developed by LignoTech. In the period
 337 2000-2009, the development and research of bio-adhesives continued, although it was not until
 338 2010 that a notable interest in this research topic could be observed (**Figure 5**). Since then, a
 339 higher number of articles has been published, with 2020 being the year that stands out with 26
 340 publications. However, the years 2017 and 2019 are equally noteworthy, with 23 and 18 articles,
 341 respectively. However, it is important to mention that, as of May 2021 (when this review was
 342 developed), a total of 8 articles had already been published, which is indicative that the strong
 343 interest in the development of adhesives from renewable natural resources will continue to
 344 grow.



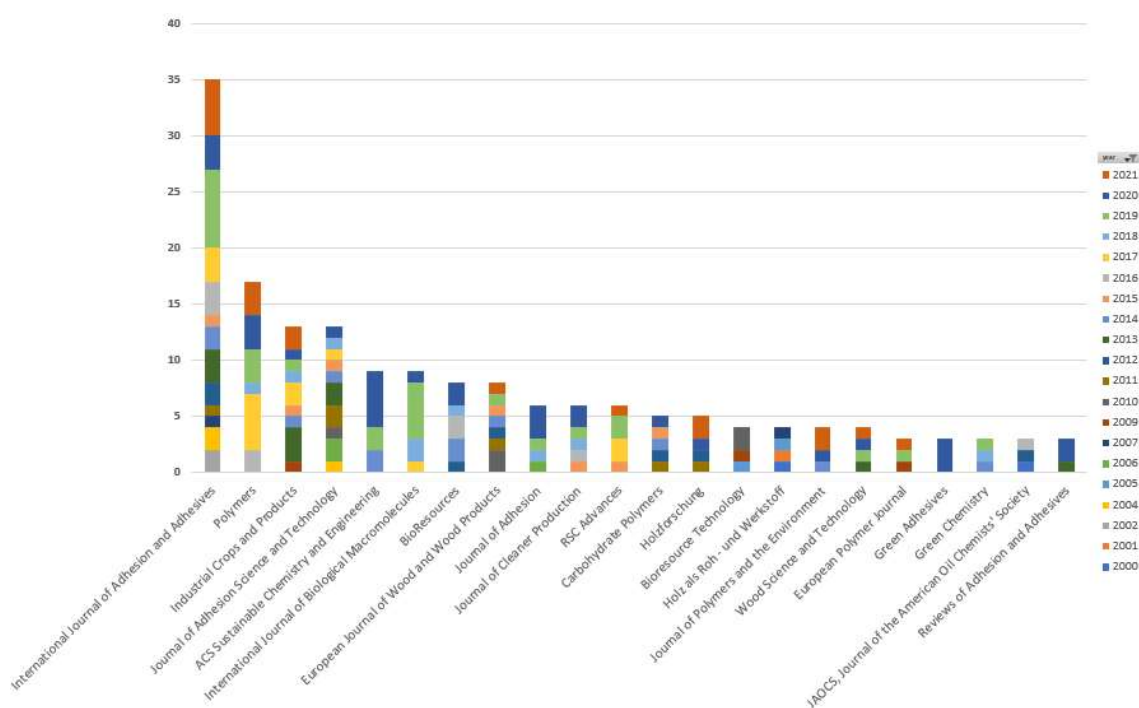
345
 346 Figure 5. Graphical diagram including the number of references published
 347 per year in the period 1999-2021.

348
 349 **4.4 Publications per Journal. Standing authors and institutions.**

350 **Figure 6** shows the journals and books with at least 3 publications. Among them, the most
 351 relevant journals that report topics on bio-adhesives are the International Journal of Adhesion
 352 and Adhesives (35 manuscripts), followed by Polymers (17 manuscripts) and Industrial Crops
 353 and Products and Journal of Adhesion Science and Technology, with 13 manuscripts published
 354 each. Regarding the top authors in terms of scientific productivity on the topic, a ranking of

355 those with more scientific publications are the following, Prof. A. Pizzi (799 SCI publications,
 356 18668 citations), Prof. J. Ling (293 SCI publications, 4259 citations), Prof. Q. Gao (164 SCI
 357 publications, 5575 citations) and Prof. J. Luo (91 SCI publications, 2248 citations).

358 Regarding the universities or institutions with higher impact on the topic: University of Lorraine
 359 and Université de Pau et des Pays de l'Adour, both located in France, together with the Beijing
 360 Forestry University and the Fujian University of Agriculture and Forestry, are the leading
 361 institutions in this research topic.



362

363

Figure 6. Number of publications per journal and year.

364

365 **5 Typology of bio-adhesives according to the bio-based materials used in their formulation.**

366 In this section, a detailed analysis of each of the most commonly used renewable-based raw
 367 materials for the development of new bio-adhesive alternatives is developed, in accordance
 368 with the results obtained after the evaluation of the bibliographic references. The different
 369 methods used for their activation / functionalization are named, as well as the current trends
 370 regarding the bio-adhesive formulation processes.

371 **5.1 Lignin-based bio-adhesives**

372 Lignin is considered to be the most abundant polymer in nature, just after cellulose (Kögel-
373 Knabner and Amelung, 2013). It is obtained as a by-product of the paper pulp industry, which
374 world production is at levels of 50-75 million tons (Antov et al., 2020), which is mainly destined
375 for energy or directly managed as waste. Aiming to increase the useful value of technical lignins
376 (lignosulfonates, organosolv lignin, or kraft lignin), the development of adhesives for wood-
377 based panels has become a high potential option (Ferdosian et al., 2017; Norström et al., 2018;
378 Papadopoulou et al., 2017; Pizzi, 2006; Solt et al., 2019; Yotov et al., 2017). The simplest way to
379 use lignin in the form of wood-based panel adhesives is based on partial substitution of phenol
380 in PF resins (Hemmila et al., 2013).

381 Although the phenolic structure of lignin provides high hydrophobicity and low dispersion
382 properties for the development of bio-adhesives, its aromatic structure decreases its reactivity,
383 so the development of lignin-based adhesives requires previous stages of functionalization of
384 the lignin structure through phenolic and aliphatic groups as well as aromatic rings (Bertella and
385 Luterbacher, 2020). The most widespread methods for modification of lignin reactivity are
386 glyoxalation (Van Nieuwenhove et al., 2020), hydroxymethylation, phenolation, demethylation
387 and oxidation reactions, among others (Hemmila et al., 2013).

388 Glyoxal is a non-toxic aldehyde that is less reactive than formaldehyde. Its low toxicity and non-
389 volatile properties make it a compound with great potential to replace formaldehyde in the
390 formulation of wood-based panel adhesives ((Mansouri et al., 2011); Tupa Esfandiyari et al.,
391 2020; Van Nieuwenhove et al., 2020). Although glyoxylation is one of the most widely used
392 activation methods for lignin functionalization, the environmental profile associated with the
393 process poses limitations to be taken into account: required concentration of glyoxal, reaction
394 time, and temperature (Navarrete et al., 2010), which implies significant energy costs. The LCA
395 of a bioadhesive for wood composed of glyoxylated lignin and polyethylenimine showed that

396 the lignin functionalization stage is the one that contributes to the greatest environmental
397 impacts, with energy requirements being the main hot spot of the process (Arias et al., 2020).
398 Thus, the development of other lignin activation processes that represent lower impacts is
399 necessary.

400 Hydroxymethylation is based on the introduction of an hydroxymethyl group (-CH₂OH) into the
401 molecular structure of lignin, specifically into the aromatic rings, in an alkaline medium (Hu et
402 al., 2011). The phenolation method developed by the addition of phenol into the aromatic side
403 chains of lignin allows enhancing and improving reactivity through phenolic hydroxyl groups in
404 the lignin structure (F. Zhang et al., 2019). Both methods, methylation and phenolation, are
405 the ones that have reached a high interest for industrial processing, due to their low cost
406 (Hemmila, et al., 2013).

407 The main goal of the lignin demethylation process is based on the conversion of the methoxyl
408 group into an hydroxyl group (M Sain, 2013), achieving higher curing rate in the wood panel
409 manufacturing process, and also improving bond strength (Li et al., 2016). Finally, regarding
410 oxidation reactions, the epoxidation stands out (Eraghi Kazzaz et al., 2019). It is characterized by
411 reacting the hydroxyl groups that make up the lignin structure with epichlorohydrin, being a
412 feasible procedure to include epoxy groups, leading to an improvement in the mechanical
413 properties of lignin and an increase in its compatibility with that of synthetic resins (Gouveia et
414 al., 2020).

415 The most recent studies on lignin-based bio-adhesives focus on the modification of the lignin
416 structure by oxidation reactions, using sodium periodate to demethylate the lignin polymeric
417 structure, which leads to an increase in the number of aromatic hydroxyl groups and the
418 reactivity of lignin (Chen et al., 2020).

419 Another trend that is gaining ground in this field is the use of lignin as a crosslinking agent for
420 the formulation of polyurethane-based wood adhesives after lignin dissolution in polypropylene

421 glycol. Then, isocyanates or polyols would be added to formulate an adhesive with a slower
422 curing time and a better bond strength (Gadhavé et al., 2019b). In addition, the formulation of
423 lignin-based adhesives will largely depend on the pretreatment steps to recover the lignin
424 (Dababi et al., 2016), thus an alkali process followed by a steam explosion will result in better
425 adhesive properties (He et al., 2020).

426 **5.2 Tannins-based bio-adhesives**

427 In addition to lignin, another renewable lignocellulosic feedstock that could be used for the
428 formulation of bio-adhesives are tannins (Zhou and Du, 2020). Tannins can be classified into two
429 groups, those characterized by having a molecular structure composed of flavonoids, known as
430 condensed tannins, and those consisting of gallic and ellagic acids with a sugar core, known as
431 hydrolysable tannins. Among these categories, condensed tannins are the most relevant with
432 90% of the world tannin market, that accounts for an annual production of 2,000,000 tons per
433 year (Das et al., 2020).

434 Tannins are obtained from extraction processes, generally hot water extraction, although other
435 solvents can also be used: acetone (Meng et al., 2019) , methanol (Romero et al., 2020), ethanol
436 (Rhazi et al., 2019), as well as sodium sulfite (Poaty et al., 2010) and NaOH (Guo et al., 2020).
437 Thus, there is a wide field of research regarding this by-product for the development of bio-
438 adhesives for wood-based panels, due to its phenolic structure and the presence of aromatic
439 rings of resorcinol that make tannins highly reactive molecules. In fact, when compared to the
440 reactivity of phenol, used for the formulation of synthetic phenol-formaldehyde (PF) adhesives,
441 its reactivity is up to 10 to 50 times higher (Pizzi, 2009). Other reasons that provide advantages
442 in the use of tannins in the processing of wood products is that they provide good strength
443 properties and increase the water resistance of wood panels (Li et al., 2019; Yang et al., 2020).
444 In addition, it promotes better wood preservation by reducing the proliferation of fungi and

445 termites, while improving dimensional stability (Tomak and Gonultas, 2018; Yalcin and Ceylan,
446 2017).

447 The main drawback in the use of tannins for the formulation of bio-based adhesives lies in their
448 high viscosity, given the existence of hydrogen bonds and electrostatic interactions in their
449 molecular structure, in addition to their high molecular weight. To reduce it, different methods
450 have been proposed (Zhou and Du, 2020), among which acid or alkaline hydrolysis with
451 aldehydes stands out, where the use of hexamethylenetetramine (or hexamine) as a hardening
452 agent is the most widespread (Moubarik et al., 2010; Pichelin et al., 1999). As in the case of
453 lignin, glyoxal is also used as a crosslinking agent (Ballerini et al., 2005). However, with the aim
454 of avoiding the need to add hardeners to the molecular structure of tannins, self-condensation
455 alternatives have been considered (Böhm et al., 2016; Dababi et al., 2020; Osman, 2013, 2012),
456 which would provide a much more environmentally friendly adhesive to be used in the
457 manufacture of interior wood panels.

458 Current experimental studies have developed bio-based adhesives using tannins as crosslinking
459 agents for soy-based adhesives, with the aim of increasing their water resistance. The phenolic
460 structure of tannins gives them a high reactivity, similar to that obtained with phenol (Ghahri et
461 al., 2021). In addition to the combination of tannins with soy, there are also recent studies
462 proposing their use for the formulation of lignin-tannin bio-adhesives, which have been shown
463 to achieve adequate shear strength (Saražin et al., 2020). On the other hand, one of the main
464 drawbacks of tannin-based bio-adhesives is their low water resistance, which reduces their
465 potential for use in the wood-based panel industry. Achieving a breakthrough with respect to
466 this property would mean a wider range of application, as it would expand their use not only for
467 the development of boards for interior use, but also for outdoor use. The research study
468 developed by (Zhang et al., 2019) has demonstrated an increase in the water resistance of a

469 tannin-based bio-adhesive by using furfuryl alcohol, glyoxal and epoxy resin as crosslinking
470 agents under acidic conditions.

471 **5.3 Soy-based bio-adhesives**

472 Several studies have demonstrated the great viability of soy to be used as a renewable raw
473 material for the development of wood bio-adhesives (Frihart et al., 2010; Frihart and Satori,
474 2013; Schwarzkopf et al., 2010; Wescott et al., 2006) to be applied in the manufacture of
475 plywood, particleboard and even medium density fiberboard (MDFB). Its potential in this
476 productive area is the result of a set of advantageous conditions that place it in a dominant
477 position for the development of new products with high added value in the market. These
478 advantages include its low cost, ease of processing, wide availability, in addition to the process
479 conditions that its use requires, i.e., low pressure and low curing temperature, factors of high
480 relevance in the process of gluing wood-based panels (Ghahri et al., 2018). However, although
481 the use of soy provides a number of advantageous properties for the development of bio-
482 adhesives, it has the disadvantage that chemical or physical modifications are required to
483 increase the water resistance and strength of the boards. For this, it is necessary to favor the
484 formation of strong wood-adhesive bonds, and the molecular structure of soy, composed of
485 several functional groups (amino, carboxylic acids, hydroxyl groups), is completely folded
486 (Vnučec et al., 2017). A process of modification and structural attack is required to favor the
487 unfolding of the complex structure that make up the soy protein, in this way the formation of
488 bonds with crosslinking agents and with wood structural complexes will be simpler, faster and
489 will provide better properties and better adhesion on wood boards (Ghahri et al., 2018).

490 To promote soy protein adhesion, the molecules have to be dispersed and denatured in solution,
491 in fact, previous studies have shown that denaturation of soy proteins leads to an increase in
492 adhesive bonding strength and water resistance (Frihart and Birkeland, 2014). Accessibility and
493 surface reactivity of soy can be achieved by chemical, thermal or enzymatic modifications. The

494 most widespread are chemical-based processes (Vnučec et al., 2017), which could be classified
495 as: denaturation by breaking the internal structure (Lin et al., 2012; Qi et al., 2013), grafting with
496 cross-linker agents (Qi et al., 2013; Zhu and Damodaran, 2014) and mixing with both bio-based
497 materials (such as lignin and tannin) or synthetic adhesives (i.e. PF, MF and MUF resins) (Lei et
498 al., 2014; Li et al., 2013, 2009; Meng et al., 2019). As for the grafting procedure, a wide variety
499 of crosslinking agents have been studied, including diacyandiamide (Solt et al., 2019), SDBS
500 (sodium dodecylbenzene sulfonate) (Lei et al., 2014), waterborne polyurethane (Wang et al.,
501 2018), PAM (polyacrylamide) (Kang et al., 2019) and maleic anhydride (Xi et al., 2020), among
502 others.

503 Arias et al., (2021) have evaluated the environmental profile of different grafting alternatives
504 for soy-based adhesives, using the crosslinking agents mentioned above. The results obtained
505 demonstrate the high potential for large-scale development of bio-adhesives for wood based on
506 this raw material, concluding that the bio-adhesive formulated with the SDBS cross-linker is the
507 one with which the lowest impact values have been obtained, from the point of view of the
508 categories of ecosystem quality, resource scarcity, and human health.

509 According to the latest research on soy-based bio-adhesives (Alcock and Peijs, 2013), the
510 concept of "self-crosslinking" and "self-reinforced composites" are being investigated to
511 improve mechanical properties by reinforcing the soy polymeric matrix with isolated soy protein
512 fibers (Kang et al., 2019). Another recent and sustainable way to improve the mechanical
513 properties of soy-based bio-adhesives is by reacting bio-epoxy bark resins with the reactive
514 groups of soy proteins. This reaction is characterized by epoxidation, thus forming a strong
515 network with improved properties of thermal resistance, strength and water resistance. In
516 addition, the use of the bark bio-epoxy resin results in higher solids content and lower viscosity
517 of the final bio-adhesive (Luo et al., 2020)

518 Finally, it is important to mention that soy-based adhesives are the ones that have achieved a
519 greater presence in the market. There are currently different companies developing adhesives
520 based on this raw material, as is the case of Soy protein-Kymene[®], an adhesive used for plywood
521 composites, which results in wood products with even higher shear strength than those glued
522 with UF resins (Vnučec et al., 2017), SOYBABY[®] and OZERO[®], adhesives developed to be applied
523 on MDF and HDF wood boards, Soyad[™] and Prolia[™] from Cargill, with extended application on
524 particleboard and plywood.

525 **5.4 Starch-based bio-adhesives**

526 Regarding starch, it is an abundant and low-cost natural polymer found in cereals (i.e., corn and
527 wheat), tubers and roots (Monteiro et al., 2016). Its molecular structure is made up of two high
528 molecular weight compounds, amylose and amylopectin. The content of these polysaccharides
529 affects the bonding strength of the starch bio-adhesives, since the higher the amount of
530 amylopectin, the higher the bonding strength (Tratnik et al., 2020; Zhou et al., 2012). Thus,
531 starch source selection is a key factor when developing starch bio-adhesives for wood bonding.
532 Nevertheless, the large amount of hydroxyl groups found in the molecular structure of starch
533 leads to an excellent affinity with polar compounds, such as cellulose (Monteiro et al., 2016).
534 This fact allows the formation of strong adhesive-wood bonds during board gluing processes,
535 but, at the same time, the simplicity of formation of hydrogen bonds by starch hydroxyl groups
536 with water molecules leads to poor water resistance and slow drying (Sun et al., 2018). These
537 mentioned shortcomings, in addition to their lower stability compared to that achieved by
538 synthetic adhesives, requires the improvement of starch bio-adhesives performance to be
539 competitive with petrochemical resins (Jiang et al., 2019). Another feature that must be taking
540 into account when considering starch as raw material for the development of wood bio-
541 adhesives is its high viscosity, derived from the macromolecules present on its molecular
542 structure, which is also a key factor when evaluating the performance and properties of the

543 bio-adhesives. In this way, it has been determined that adding small molecules into the
544 formulation of the starch bio-adhesive, like for example low molecular weight crosslinking
545 agents, is an adequate and valuable procedure to decrease the entanglements, leading to
546 greater fluidity and easier penetration into the surface of the wood (Din et al., 2020).

547 Although there are several methods by which the starch structure can be modified (i.e., physical,
548 chemical, and enzymatic), the ones that stand out are the chemical ones, specifically graft
549 copolymerization, as it is considered one of the best ways to increase the adhesion properties
550 (Gu et al., 2019; V. Gadhave et al., 2017). Different crosslinking agents have been proposed to
551 develop the copolymerization of the starch molecular structure, with hexamethoxymethyl-
552 melamine (Sridach et al., 2013), polyvinyl acetate (Nie et al., 2013), pMDI (polymeric-methylene
553 diphenyl-diisocyanate) (Gu et al., 2010) and glyoxal (Deng et al., 2018; Gadhave et al., 2019a)
554 being the most widely used.

555 Another widespread method is oxidation, in which the hydroxyl groups located at the C-2, C-3
556 and C-6 positions of the starch carbon chains are transformed into carboxylic and carbonyl
557 groups (Sun et al., 2018). The oxidation of starch with hydrogen peroxide has been previously
558 investigated, as this compound decomposes into hydrogen ions and water, leading to low
559 environmental impact (Zhang et al., 2015). The main drawback of this procedure is the high
560 reactivity of the carboxyl and carbonyl groups, which leads to further cross-linking reactions,
561 resulting in low bonding strength and low water resistance (Zhang et al., 2015). Therefore, to
562 increase the technical performance of starch bio-adhesives, a second process step, consisting of
563 graft copolymerization with SDS (sodium dodecyl sulfate) (Li et al., 2014), silane coupling agents
564 or olefin monomers (De Bruyn et al., 2007; Tanrattanakul and Chumeka, 2010), among others,
565 is required. By this addition, both drawbacks (bonding strength and water resistance) are
566 significantly improved (Zhang et al., 2015).

567 Finally, another trend that is gaining ground in the field of study of starch-based bio-adhesives
568 is the use of citric acid in their formulation (Amini et al., 2020; Kusumah et al., 2020; Kusumah
569 et al., 2017; Umemura et al., 2012), which allowed to achieve standard-compliant mechanical
570 and physical properties (Kusumah et al., 2020). The formulation of the bio-adhesive requires a
571 previous chemical modification of the starch, which is based on a first acid hydrolysis, to obtain
572 maltodextrin, which is able to react with the hydroxyl groups present in the wood, leading to
573 the formation of high strength bonds (ester and glycoside bonds) between the bio-adhesive and
574 the wood board.

575 **6. Conclusions**

576 This paper provided a critical analysis of high impact literature on bio-based adhesives to be
577 used for the manufacture of wood-based panels. A large number of experimental studies were
578 observed, whereas only a few literature reviews and patents were found. The development of
579 patents is not extensive, which, therefore, is left as a recommendation in order to aid industrial
580 development.

581 Among all the raw materials for the formulation of bio-adhesives, soybean seems to be the one
582 that stands out, and has achieved a greater presence in the market. It has been observed that
583 companies have been developing adhesives based on this raw material, and such practice seems
584 to be a positive trend in sustainable terms.

585 There is evidence that this body of literature (bio-based adhesives for the manufacture of wood-
586 based panels) has been increasingly receiving contributions globally, and the number of
587 publications in the area has been increasing. Since 2010 a greater number of publications can
588 be observed, with 26 publications having been observed in the year 2020. This is indicative that
589 the great interest in the development of adhesives from renewable natural resources will
590 continue to grow. In addition, the most prominent journals publishing research involving bio-

591 adhesives are the *International Journal of Adhesion and Adhesives*, with 26 documents, and
592 *Polymers*, with 13.

593 The required data on energy and electrical consumption, process equipment, and emissions to
594 the environment, are not currently available in the existing literature, therefore expanding the
595 portfolio of studies may bring more consistency to the results, mainly for life cycle assessments.
596 Drawing on the limitations of this research, the authors neither claim it to be exempt from
597 limitations nor exhaustive, as it was limited to the particular keywords and databases presented
598 in the methods section. Nonetheless, the authors believe the portfolio of documents identified
599 and used in this study to be representative of the body of literature on bio-based adhesives for
600 the manufacture of wood-based panels.

601 The critical analysis developed in this review is timely not only for researchers but also for
602 professionals working in R&D areas of the productive sector. The conclusions derived from each
603 bio-adhesive typology serve as a guide for decision-making for future developments and
604 practical applications. It would be ideal that in the coming years a greater number of
605 environmental studies on the different wood bio-adhesives would be carried out because,
606 although most experimental studies on bio-adhesives report that they are environmentally
607 friendly alternatives, there are limited studies that have investigated the production routes from
608 a global point of view and that, therefore, endorse this label from a holistic, objective and
609 quantitative point of view.

610 **Funding**

611 This research has been financially supported by ERA-CoBIOTECH project (PCI2018-092866)
612 Programación Conjunta Internacional 2018-WOODBADH project. S. Ana Arias, S. González-
613 Rodríguez and Prof. María Teresa Moreira belong to CRETUS and the Galician Competitive
614 Research Group GRC ED431C 2017/29, co-founded by Xunta de Galicia and FEDER (EU). S.
615 González-Rodríguez thanks to the Spanish Ministry of Science, Innovation and Universities for

616 financial support (Grant reference BES-2017-081677). Authors Murillo Vetroni Barros and
617 Rodrigo Salvador have received research grants from the *Coordenação de Aperfeiçoamento de*
618 *Pessoal de Nível Superior - Brasil* (CAPES) - Finance Code 001. Authors Antonio Carlos de
619 Francisco and Cassiano Moro Piekarski have received research grants from the *Conselho*
620 *Nacional de Desenvolvimento Científico e Tecnológico* (CNPq) (Sponsored by CNPq
621 310686/2017-2 and 312285/2019-1).

622

623

624

625

626

627

628

629

630

631

632

633

634

635

636

637 **References**

- 638 Alcock, B., Peijs, T., 2013. Technology and development of self-reinforced polymer composites.
639 Adv. Polym. Sci. 251. https://doi.org/10.1007/12_2011_159
- 640 Amini, M.H.M., Hashim, R., Sulaiman, N.S., Mohamed, M., Sulaiman, O., 2020. Citric acid-
641 modified starch as an environmentally friendly binder for wood composite making.
642 BioResources 15, 4234–4248. <https://doi.org/10.15376/biores.15.2.4234-4248>
- 643 Antov, P., Mantanis, G.I., Savov, V., 2020. Development of wood composites from recycled fibres
644 bonded with magnesium lignosulfonate. Forests. <https://doi.org/10.3390/F11060613>
- 645 Antov, P., Savov, V., Krišt'ák, L., Réh, R., Mantanis, G.I., 2021. Eco-friendly, high-density
646 fiberboards bonded with urea-formaldehyde and ammonium lignosulfonate. Polymers
647 (Basel). <https://doi.org/10.3390/polym13020220>
- 648 Arias, A., González-García, S., Feijoo, G., Moreira, M.T., 2021. Environmental benefits of soy-
649 based bio-adhesives as an alternative to formaldehyde-based options. Environ. Sci. Pollut.
650 Res. 1–14. <https://doi.org/10.1007/s11356-021-12766-4>
- 651 Arias, A., González-García, S., González-Rodríguez, S., Feijoo, G., Moreira, M.T., 2020. Cradle-to-
652 gate Life Cycle Assessment of bio-adhesives for the wood panel industry. A comparison
653 with petrochemical alternatives. Sci. Total Environ. <https://doi.org/10.1016/j.scitotenv.2020.140357>
- 655 Aziz, N.A., Latip, A.F.A., Peng, L.C., Latif, N.H.A., Brosse, N., Hashim, R., Hussin, M.H., 2019.
656 Reinforced lignin-phenol-glyoxal (LPG) wood adhesives from coconut husk. Int. J. Biol.
657 Macromol. <https://doi.org/10.1016/j.ijbiomac.2019.08.255>
- 658 Ballerini, A., Despres, A., Pizzi, A., 2005. Non-toxic, zero emission tannin-glyoxal adhesives for
659 wood panels. Holz als Roh - und Werkst. <https://doi.org/10.1007/s00107-005-0048-x>
- 660 Bertella, S., Luterbacher, J.S., 2020. Lignin Functionalization for the Production of Novel
661 Materials. Trends Chem. <https://doi.org/10.1016/j.trechm.2020.03.001>
- 662 Böhm, R., Hauptmann, M., Pizzi, A., Friedrich, C., Laborie, M.P., 2016. The chemical, kinetic and
663 mechanical characterization of tannin-based adhesives with different crosslinking systems.
664 Int. J. Adhes. Adhes. 68. <https://doi.org/10.1016/j.ijadhadh.2016.01.006>
- 665 Chen, N., Zheng, P., Zeng, Q., Lin, Q., Rao, J., 2017. Characterization and performance of soy-

666 based adhesives cured with epoxy resin. *Polymers* (Basel).
667 <https://doi.org/10.3390/polym9100514>

668 Chen, X., Xi, X., Pizzi, A., Fredon, E., Du, G., Gerardin, C., Amirou, S., 2020. Oxidized demethylated
669 lignin as a bio-based adhesive for wood bonding. *J. Adhes.*
670 <https://doi.org/10.1080/00218464.2019.1710830>

671 Dababi, I., Gimello, O., Elaloui, E., Brosse, N., 2020. Water Extraction of Tannins from Aleppo
672 Pine Bark and Sumac Root for the Production of Green Wood Adhesives. *Molecules* 25.
673 <https://doi.org/10.3390/molecules25215041>

674 Dababi, I., Gimello, O., Elaloui, E., Quignard, F., Brosse, N., 2016. Organosolv lignin-based wood
675 adhesive. Influence of the lignin extraction conditions on the adhesive performance.
676 *Polymers* (Basel). <https://doi.org/10.3390/polym8090340>

677 Das, A.K., Islam, M.N., Faruk, M.O., Ashaduzzaman, M., Dungani, R., 2020. Review on tannins:
678 Extraction processes, applications and possibilities. *South African J. Bot.*
679 <https://doi.org/10.1016/j.sajb.2020.08.008>

680 De Bruyn, H., Sprong, E., Gaborieau, M., Roper, J.A., Gilbert, R.G., 2007. Starch-graft-(synthetic
681 copolymer) latexes initiated with Ce⁴⁺ and stabilized by amylopectin. *J. Polym. Sci. Part A*
682 *Polym. Chem.* 45. <https://doi.org/10.1002/pola.22189>

683 Deng, S., Pizzi, A., Du, G., Lagel, M.C., Delmotte, L., Abdalla, S., 2018. Synthesis, structure
684 characterization and application of melamine–glyoxal adhesive resins. *Eur. J. Wood Wood*
685 *Prod.* 76. <https://doi.org/10.1007/s00107-017-1184-9>

686 Din, Z. ud, Chen, L., Xiong, H., Wang, Z., Ullah, I., Lei, W., Shi, D., Alam, M., Ullah, H., Khan, S.A.,
687 2020. Starch: An Undisputed Potential Candidate and Sustainable Resource for the
688 Development of Wood Adhesive. *Starch/Staerke*. <https://doi.org/10.1002/star.201900276>

689 El Mansouri, N.E., Pizzi, A., Salvadó, J., 2007. Lignin-based wood panel adhesives without
690 formaldehyde. *Holz als Roh - und Werkst.* <https://doi.org/10.1007/s00107-006-0130-z>

691 Eraghi Kazzaz, A., Hosseinpour Feizi, Z., Fatehi, P., 2019. Grafting strategies for hydroxy groups
692 of lignin for producing materials. *Green Chem.* <https://doi.org/10.1039/c9gc02598g>

693 Eslah, F., Jonoobi, M., Faezipour, M., Afsharpour, M., Enayati, A.A., 2016. Preparation and
694 development of a chemically modified bio-adhesive derived from soybean flour protein.
695 *Int. J. Adhes. Adhes.* <https://doi.org/10.1016/j.ijadhadh.2016.08.011>

696 Ferdosian, F., Pan, Z., Gao, G., Zhao, B., 2017. Bio-based adhesives and evaluation for wood
697 composites application. *Polymers (Basel)*. <https://doi.org/10.3390/polym9020070>

698 França, W.T., Barros, M.V., Salvador, R., de Francisco, A.C., Moreira, M.T., Piekarski, C.M., 2021.
699 Integrating life cycle assessment and life cycle cost: a review of environmental-economic
700 studies. *Int. J. Life Cycle Assess.* <https://doi.org/10.1007/s11367-020-01857-y>

701 Frihart, C.R., Birkeland, M.J., 2014. Soy properties and soy wood adhesives, in: ACS Symposium
702 Series. <https://doi.org/10.1021/bk-2014-1178.ch008>

703 Frihart, C.R., Birkeland, M.J., Allen, A.J., Wescott, J.M., 2010. Soy Adhesives that Can Form
704 Durable Bonds for Plywood , Laminated Wood Flooring , and Particleboard. *Proc. Int. Conv.*
705 *Soc. Wood Sci. Technol. United Nations Econ. Comm. Eur. – Timber Comm.* Oct. 11-14,
706 2010, Geneva, Switz.

707 Frihart, C.R., Satori, H., 2013. Soy flour dispersibility and performance as wood adhesive. *J.*
708 *Adhes. Sci. Technol.* 27. <https://doi.org/10.1080/01694243.2012.696948>

709 Gadhave, R. V., Mahanwar, P.A., Gadekar, P.T., 2019a. Study on Various Compositions of
710 Polyvinyl Alcohol and Starch Blends by Cross-Linking with Glyoxal. *Open J. Polym. Chem.*
711 09. <https://doi.org/10.4236/ojchem.2019.94007>

712 Gadhave, R. V., S. Kasbe, P., Mahanwar, P.A., Gadekar, P.T., 2019b. Synthesis and
713 characterization of lignin-polyurethane based wood adhesive. *Int. J. Adhes. Adhes.* 95.
714 <https://doi.org/10.1016/j.ijadhadh.2019.102427>

715 Ghahri, S., Chen, X., Pizzi, A., Hajihassani, R., Papadopoulos, A.N., 2021. Natural tannins as new
716 cross-linking materials for soy-based adhesives. *Polymers (Basel)*. 13, 1–15.
717 <https://doi.org/10.3390/polym13040595>

718 Ghahri, S., Mohebbi, B., Pizzi, A., Mirshokraie, A., Mansouri, H.R., 2018. Improving Water
719 Resistance of Soy-Based Adhesive by Vegetable Tannin. *J. Polym. Environ.* 26.
720 <https://doi.org/10.1007/s10924-017-1090-6>

721 Gouveia, J.R., Garcia, G.E.S., Antonino, L.D., Tavares, L.B., Dos Santos, D.J., 2020. Epoxidation of
722 Kraft Lignin as a Tool for Improving the Mechanical Properties of Epoxy Adhesive.
723 *Molecules*. <https://doi.org/10.3390/molecules25112513>

724 Gu, J., Zuo, Y., Zhang, Y., Tan, H., Zhu, L., Shen, J., 2010. Preparation of plywood using starch
725 adhesives modified with isocyanate, in: *Applied Mechanics and Materials*.

726 <https://doi.org/10.4028/www.scientific.net/AMM.26-28.1065>

727 Gu, Y., Cheng, L., Gu, Z., Hong, Y., Li, Z., Li, C., 2019. Preparation, characterization and properties
728 of starch-based adhesive for wood-based panels. *Int. J. Biol. Macromol.*
729 <https://doi.org/10.1016/j.ijbiomac.2019.04.088>

730 Guo, L., Qiang, T., Ma, Y., Wang, K., Du, K., 2020. Optimisation of tannin extraction from *Coriaria*
731 *nepalensis* bark as a renewable resource for use in tanning. *Ind. Crops Prod.* 149.
732 <https://doi.org/10.1016/j.indcrop.2020.112360>

733 He, Q., Ziegler-Devin, I., Chrusciel, L., Obame, S.N., Hong, L., Lu, X., Brosse, N., 2020. Lignin-First
734 Integrated Steam Explosion Process for Green Wood Adhesive Application. *ACS Sustain.*
735 *Chem. Eng.* 8, 5380–5392. <https://doi.org/10.1021/acssuschemeng.0c01065>

736 Head, M., Levasseur, A., Beauregard, R., Margni, M., 2020. Dynamic greenhouse gas life cycle
737 inventory and impact profiles of wood used in Canadian buildings. *Build. Environ.* 173.
738 <https://doi.org/10.1016/j.buildenv.2020.106751>

739 Hemmila, V; Trischler, J; Sandberg, D., 2013. LIGNIN – AN ADHESIVE RAW MATERIAL OF THE
740 FUTURE OR WASTE OF RESEARCH ENERGY ? *Sci. Eng.*

741 Hosseinpourpia, R., Adamopoulos, S., Mai, C., Taghiyari, H.R., 2019. Properties of medium-
742 density fibreboards bonded with dextrin-based wood adhesive. *Wood Res.* 64.

743 Hu, L., Pan, H., Zhou, Y., Zhang, M., 2011. Methods to improve lignin’s reactivity as a phenol
744 substitute and as replacement for other phenolic compounds: A brief review.
745 *BioResources.* <https://doi.org/10.15376/biores.6.3.3515-3525>

746 Jiang, Y., Chen, Q., Tan, H., Gu, J., Zhang, Y., 2019. A low-cost, formaldehyde-free, and high-
747 performance starch-based wood adhesive. *BioResources* 14.
748 <https://doi.org/10.15376/biores.14.1.1405-1418>

749 Kang, H., Wang, Z., Wang, Y., Zhao, S., Zhang, S., Li, J., 2019. Development of mainly plant
750 protein-derived plywood bioadhesives via soy protein isolate fiber self-reinforced soybean
751 meal composites. *Ind. Crops Prod.* <https://doi.org/10.1016/j.indcrop.2019.03.022>

752 Kirk-Othmer, 1998. *Kirk-Othmer Encyclopedia of Chemical Technology - Vol 8.* J. Am. Chem. Soc.

753 Kögel-Knabner, I., Amelung, W., 2013. Dynamics, Chemistry, and Preservation of Organic Matter
754 in Soils, in: *Treatise on Geochemistry: Second Edition.* [https://doi.org/10.1016/B978-0-08-](https://doi.org/10.1016/B978-0-08-095975-7.01012-3)
755 [095975-7.01012-3](https://doi.org/10.1016/B978-0-08-095975-7.01012-3)

- 756 Kusumah, S.S., Jayadi, Wibowo, D.T., Pramasari, D.A., Widyaningrum, B.A., Darmawan, T.,
757 Ismadi, Dwianto, W., Umemura, K., 2020. Investigation of eco-friendly plywood bonded
758 with citric acid - Starch based adhesive, in: IOP Conference Series: Earth and Environmental
759 Science. Institute of Physics Publishing. <https://doi.org/10.1088/1755-1315/460/1/012009>
- 760 Kusumah, S.S., Umemura, K., Guswenrivo, I., Yoshimura, T., Kanayama, K., 2017. Utilization of
761 sweet sorghum bagasse and citric acid for manufacturing of particleboard II: influences of
762 pressing temperature and time on particleboard properties. *J. Wood Sci.* 63.
763 <https://doi.org/10.1007/s10086-016-1605-0>
- 764 Kusumah, S.S., Umemura, K., Yoshioka, K., Miyafuji, H., Kanayama, K., 2016. Utilization of sweet
765 sorghum bagasse and citric acid for manufacturing of particleboard I: Effects of pre-drying
766 treatment and citric acid content on the board properties. *Ind. Crops Prod.* 84.
767 <https://doi.org/10.1016/j.indcrop.2016.01.042>
- 768 Lei, H., Du, G., Wu, Z., Xi, X., Dong, Z., 2014. Cross-linked soy-based wood adhesives for plywood.
769 *Int. J. Adhes. Adhes.* 50, 199–203. <https://doi.org/10.1016/j.ijadhadh.2014.01.026>
- 770 Li, Jiongjiong, Wang, W., Zhang, S., Gao, Q., Zhang, W., Li, Jianzhang, 2016. Preparation and
771 characterization of lignin demethylated at atmospheric pressure and its application in fast
772 curing biobased phenolic resins. *RSC Adv.* 6. <https://doi.org/10.1039/c6ra11966b>
- 773 Li, Jiongjiong, Zhu, W., Zhang, S., Gao, Q., Xia, C., Zhang, W., Li, Jianzhang, 2019.
774 Depolymerization and characterization of Acacia mangium tannin for the preparation of
775 mussel-inspired fast-curing tannin-based phenolic resins. *Chem. Eng. J.* 370.
776 <https://doi.org/10.1016/j.cej.2019.03.211>
- 777 Li, X., Chen, M., Zhang, J., Gao, Q., Zhang, S., Li, J., 2017. Physico-chemical properties of soybean
778 meal-based adhesives reinforced by ethylene glycol diglycidyl ether and modified
779 nanocrystalline cellulose. *Polymers (Basel)*. <https://doi.org/10.3390/polym9090463>
- 780 Li, X., Li, Y., Zhong, Z., Wang, D., Ratto, J.A., Sheng, K., Sun, X.S., 2009. Mechanical and water
781 soaking properties of medium density fiberboard with wood fiber and soybean protein
782 adhesive. *Bioresour. Technol.* 100. <https://doi.org/10.1016/j.biortech.2009.02.048>
- 783 Li, X., Wang, D., Ratto, J.A., Sun, X.S., 2013. Production and characterization of high strength,
784 thin-layered, pulp fiberboard using soy protein adhesives. *J. Adhes. Sci. Technol.* 27.
785 <https://doi.org/10.1080/01694243.2012.696957>
- 786 Li, Z., Wang, J., Cheng, L., Gu, Z., Hong, Y., Kowalczyk, A., 2014. Improving the performance of

787 starch-based wood adhesive by using sodium dodecyl sulfate. *Carbohydr. Polym.* 99, 579–
788 583. <https://doi.org/10.1016/j.carbpol.2013.08.062>

789 Lin, Q., Chen, N., Bian, L., Fan, M., 2012. Development and mechanism characterization of high
790 performance soy-based bio-adhesives. *Int. J. Adhes. Adhes.*
791 <https://doi.org/10.1016/j.ijadhadh.2012.01.005>

792 Luo, J., Zhou, Y., Gao, Q., Li, J., Yan, N., 2020. From Wastes to Functions: A New Soybean Meal
793 and Bark-Based Adhesive. *ACS Sustain. Chem. Eng.*
794 <https://doi.org/10.1021/acssuschemeng.0c02413>

795 M Sain, N.Y., 2013. A New Method for Demethylation of Lignin from Woody Biomass using
796 Biophysical Methods. *J. Chem. Eng. Process Technol.* 04. <https://doi.org/10.4172/2157-7048.1000160>

798 Mansouri, N.E. El, Yuan, Q., Huang, F., 2011. Study of chemical modification of alkaline lignin by
799 the glyoxalation reaction. *BioResources* 6. <https://doi.org/10.15376/biores.6.4.4523-4536>

800 Mathias, J.D., Grédiac, M., Michaud, P., 2016. Bio-based adhesives, in: *Biopolymers and Biotech*
801 *Admixtures for Eco-Efficient Construction Materials.* <https://doi.org/10.1016/B978-0-08-100214-8.00016-6>

803 Meng, J., Lin, X., Zhou, J., Zhang, R., Chen, Y., Long, X., Shang, R., Luo, X., 2019. Preparation of
804 tannin-immobilized gelatin/PVA nanofiber band for extraction of uranium (VI) from
805 simulated seawater. *Ecotoxicol. Environ. Saf.* 170.
806 <https://doi.org/10.1016/j.ecoenv.2018.11.089>

807 Monteiro, S., Martins, J., Magalhães, F.D., Carvalho, L., 2016. Low density wood-based
808 particleboards bonded with foamable sour cassava starch: Preliminary studies. *Polymers*
809 (Basel). 8. <https://doi.org/10.3390/polym8100354>

810 Moubarik, A., Charrier, B., Allal, A., Charrier, F., Pizzia, A., 2010. Development and optimization
811 of a new formaldehyde-free cornstarch and tanninwood adhesive. *Eur. J. Wood Wood*
812 *Prod.* <https://doi.org/10.1007/s00107-009-0357-6>

813 Navarrete, P., Mansouri, H.R., Pizzi, A., Tapin-Lingua, S., Benjelloun-Mlayah, B., Pasch, H.,
814 Rigolet, S., 2010. Wood panel adhesives from low molecular mass lignin and tannin without
815 synthetic resins. *J. Adhes. Sci. Technol.* 24, 1597–1610.
816 <https://doi.org/10.1163/016942410X500972>

- 817 Nie, Y., Tian, X., Liu, Y., Wu, K., Wang, J., 2013. Research on starch-g-polyvinyl acetate and epoxy
818 resin-modified corn starch adhesive. Polym. Compos. 34.
819 <https://doi.org/10.1002/pc.22379>
- 820 Norström, E., Demircan, D., Fogelström, L., Khabbaz, F., Malmström, E., 2018. Green Binders for
821 Wood Adhesives, in: Applied Adhesive Bonding in Science and Technology.
822 <https://doi.org/10.5772/intechopen.72072>
- 823 Osman, Z., 2013. Comparative Thermodynamic Study on the Contribution of the
824 Autocondensation and Copolymerization Reactions for the Tannins of the Subspecies of
825 *Acacia nilotica*. J. Polym. Environ. 21. <https://doi.org/10.1007/s10924-013-0611-1>
- 826 Osman, Z., 2012. Thermomechanical analysis of the tannins of *Acacia Nilotica* spp. *Nilotica* as a
827 rapid tool for the evaluation of wood-based adhesives. J. Therm. Anal. Calorim. 107.
828 <https://doi.org/10.1007/s10973-011-1721-4>
- 829 Pagani, R.N., Kovaleski, J.L., Resende, L.M., 2015. Methodi Ordinatio: a proposed methodology
830 to select and rank relevant scientific papers encompassing the impact factor, number of
831 citation, and year of publication. Scientometrics 105. <https://doi.org/10.1007/s11192-015-1744-x>
- 833 Papadopoulou, E., Kountouras, S., Chrissafis, K., Kirpluks, M., Cabulis, U., Švíglerová, P.,
834 Benjelloun-Mlayah, B., 2017. Evaluation of the particle size of organosolv Lignin in the
835 synthesis of resol resins for plywood and their performance on fire spreading. Tappi J.
836 <https://doi.org/10.32964/tj16.7.409>
- 837 Park, C.B., Song, M.J., Choi, N.W., Kim, Sunghoon, Jeon, H.P., Kim, Sanghun, Kim, Y., 2017. An
838 eco-safety assessment of glyoxal-containing cellulose ether on freeze-dried microbial
839 strain, cyanobacteria, *Daphnia*, and Zebrafish. Int. J. Environ. Res. Public Health 14.
840 <https://doi.org/10.3390/ijerph14030323>
- 841 Pichelin, F., Kamoun, C., Pizzi, A., 1999. Hexamine hardener behaviour: Effects on wood glueing,
842 tannin and other wood adhesives. Holz als Roh - und Werkst. 57.
843 <https://doi.org/10.1007/s001070050349>
- 844 Pizzi, A., 2009. Polyflavonoid tannins self-condensation adhesives for wood particleboard. J.
845 Adhes. 85. <https://doi.org/10.1080/00218460902781917>
- 846 Pizzi, A., 2006. Recent developments in eco-efficient bio-based adhesives for wood bonding:
847 Opportunities and issues. J. Adhes. Sci. Technol.

- 848 <https://doi.org/10.1163/156856106777638635>
- 849 Poaty, B., Dumarçay, S., Gérardin, P., Perrin, D., 2010. Modification of grape seed and wood
850 tannins to lipophilic antioxidant derivatives. *Ind. Crops Prod.* 31.
851 <https://doi.org/10.1016/j.indcrop.2010.02.003>
- 852 Pocius, A. V., 2012. Adhesives and Sealants, in: *Polymer Science: A Comprehensive Reference*,
853 10 Volume Set. <https://doi.org/10.1016/B978-0-444-53349-4.00210-7>
- 854 Popović, M., Điporovic-Momčilović, M., Gavrilović-Grmuša, I., 2020. New standards and
855 regulations on formaldehyde emission from wood-based composite panels. *Zast. Mater.*
856 61. <https://doi.org/10.5937/zasmat2002152p>
- 857 Qi, G., Li, N., Wang, D., Sun, X.S., 2013. Physicochemical properties of soy protein adhesives
858 modified by 2-octen-1-ylsuccinic anhydride. *Ind. Crops Prod.* 46.
859 <https://doi.org/10.1016/j.indcrop.2013.01.024>
- 860 Rhazi, N., Hannache, H., Oumam, M., Sesbou, A., Charrier, B., Pizzi, A., Charrier-El Bouhtoury, F.,
861 2019. Green extraction process of tannins obtained from Moroccan *Acacia mollissima*
862 barks by microwave: Modeling and optimization of the process using the response surface
863 methodology RSM. *Arab. J. Chem.* 12. <https://doi.org/10.1016/j.arabjc.2015.04.032>
- 864 Romero, R., Contreras, D., Sepúlveda, M., Moreno, N., Segura, C., Melin, V., 2020. Assessment
865 of a Fenton reaction driven by insoluble tannins from pine bark in treating an emergent
866 contaminant. *J. Hazard. Mater.* 382. <https://doi.org/10.1016/j.jhazmat.2019.120982>
- 867 Ruter, S., Hendrick, E., Pingoud, K., Grêt-Regamey, A., Hetsch, S., 2008. Challenges and
868 Opportunities of Accounting for Harvested Wood Products, in: *Background Paper to the*
869 *Workshop on Harvested Wood Products in the Context of Climate Change Policies.*
- 870 Salem, M.Z.M., Böhm, M., 2013. Understanding of formaldehyde emissions from solid wood: An
871 overview. *BioResources.* <https://doi.org/10.15376/biores.8.3.4775-4790>
- 872 Saražin, J., Schmiedl, D., Pizzi, A., Šernek, M., 2020. Bio-based adhesive mixtures of pine tannin
873 and different types of lignins. *BioResources* 15, 9401–9412.
874 <https://doi.org/10.15376/biores.15.4.9401-9412>
- 875 Schwarzkopf, M., Huang, J., Li, K., 2010. A formaldehyde-free soy-based adhesive for making
876 oriented strandboard. *J. Adhes.* 86. <https://doi.org/10.1080/00218460903482549>
- 877 Solt, P., Konnerth, J., Gindl-Altmutter, W., Kantner, W., Moser, J., Mitter, R., van Herwijnen,

878 H.W.G., 2019. Technological performance of formaldehyde-free adhesive alternatives for
879 particleboard industry. *Int. J. Adhes. Adhes.* 94, 99–131.
880 <https://doi.org/10.1016/j.ijadhadh.2019.04.007>

881 Sridach, W., Jonjankiat, S., Wittaya, T., 2013. Effect of citric acid, PVOH, and starch ratio on the
882 properties of cross-linked poly(vinyl alcohol)/starch adhesives. *J. Adhes. Sci. Technol.* 27.
883 <https://doi.org/10.1080/01694243.2012.753394>

884 Sun, J., Li, L., Cheng, H., Huang, W., 2018. Preparation, characterization and properties of an
885 organic siloxane-modified cassava starch-based wood adhesive. *J. Adhes.* 94.
886 <https://doi.org/10.1080/00218464.2016.1268958>

887 Tanrattanakul, V., Chumeka, W., 2010. Effect of potassium persulfate on graft copolymerization
888 and mechanical properties of cassava starch/natural rubber foams. *J. Appl. Polym. Sci.* 116.
889 <https://doi.org/10.1002/app.31514>

890 Tomak, E.D., Gonultas, O., 2018. The Wood Preservative Potentials of Valonia, Chestnut, Tara
891 and Sulphited Oak Tannins. *J. Wood Chem. Technol.* 38.
892 <https://doi.org/10.1080/02773813.2017.1418379>

893 Tratnik, N., Kuo, P.Y., Tanguy, N.R., Gnanasekar, P., Yan, N., 2020. Biobased Epoxidized Starch
894 Wood Adhesives: Effect of Amylopectin and Amylose Content on Adhesion Properties. *ACS*
895 *Sustain. Chem. Eng.* 8, 17997–18005. <https://doi.org/10.1021/acssuschemeng.0c05716>

896 Tupa Esfandiyari, M.R., Pour, M.T., Khademieslam, H., Mir Shokraei, S.A., Bazyar, B., 2020.
897 Investigating the Possibility of Making Lignin-glyoxal Resins as Adhesives in the Production
898 of Plywood. *BioResources* 14. <https://doi.org/10.15376/biores.14.3.7122-7133>

899 Umemura, K., Ueda, T., Munawar, S.S., Kawai, S., 2012. Application of citric acid as natural
900 adhesive for wood. *J. Appl. Polym. Sci.* 123. <https://doi.org/10.1002/app.34708>

901 V. Gadhave, R., Mahanwar, P.A., Gadekar, P.T., 2017. Starch-Based Adhesives for Wood/Wood
902 Composite Bonding: Review. *Open J. Polym. Chem.*
903 <https://doi.org/10.4236/ojpchem.2017.72002>

904 Valenzuela, J., Von Leyser, E., Pizzi, A., Westermeyer, C., Gorrini, B., 2012. Industrial production
905 of pine tannin-bonded particleboard and MDF. *Eur. J. Wood Wood Prod.* 70.
906 <https://doi.org/10.1007/s00107-012-0610-2>

907 Van Nieuwenhove, I., Renders, T., Lauwaert, J., De Roo, T., De Clercq, J., Verberckmoes, A., 2020.

- 908 Biobased Resins Using Lignin and Glyoxal. ACS Sustain. Chem. Eng.
909 <https://doi.org/10.1021/acssuschemeng.0c07227>
- 910 Vnučec, D., Kutnar, A., Goršek, A., 2017. Soy-based adhesives for wood-bonding—a review. J.
911 Adhes. Sci. Technol. <https://doi.org/10.1080/01694243.2016.1237278>
- 912 Wang, Y., Deng, L., Fan, Y., 2018. Preparation of Soy-Based Adhesive Enhanced by Waterborne
913 Polyurethane: Optimization by Response Surface Methodology. Adv. Mater. Sci. Eng. 2018.
914 <https://doi.org/10.1155/2018/9253670>
- 915 Wescott, J.M., Frihart, C.R., Traska, A.E., 2006. High-soy-containing water-durable adhesives. J.
916 Adhes. Sci. Technol. 20. <https://doi.org/10.1163/156856106777638734>
- 917 Wilson, J.B., 2010. Life-cycle inventory of formaldehyde-based resins used in wood composites
918 in terms of resources, emissions, energy and carbon. Wood Fiber Sci. 42.
- 919 Xi, X., Pizzi, A., Gerardin, C., Chen, X., Amirou, S., 2020. Soy protein isolate-based polyamides as
920 wood adhesives. Wood Sci. Technol. <https://doi.org/10.1007/s00226-019-01141-9>
- 921 Yalcin, M., Ceylan, H., 2017. The effects of tannins on adhesion strength and surface roughness
922 of varnished wood after accelerated weathering. J. Coatings Technol. Res. 14.
923 <https://doi.org/10.1007/s11998-016-9841-1>
- 924 Yang, T., Dong, M., Cui, J., Gan, L., Han, S., 2020. Exploring the formaldehyde reactivity of tannins
925 with different molecular weight distributions: bayberry tannins and larch tannins.
926 Holzforschung 74. <https://doi.org/10.1515/hf-2019-0050>
- 927 Yotov, N., Valchev, I., Petrin, S., Savov, V., 2017. Lignosulphonate and waste technical hydrolysis
928 lignin as adhesives for eco-friendly fiberboard. Bulg. Chem. Commun. (9th Natl. Conf.
929 Chem. Sofia, Bulg. Sep.29 - Oct.01, 2016).
- 930 Zhang, F., Jiang, X., Lin, J., Zhao, G., Chang, H.M., Jameel, H., 2019. Reactivity improvement by
931 phenolation of wheat straw lignin isolated from a biorefinery process. New J. Chem. 43.
932 <https://doi.org/10.1039/c8nj05016c>
- 933 Zhang, J., Xi, X., Liang, J., Pizzi, A., Du, G., Deng, S., 2019. Tannin-based adhesive cross-linked by
934 furfuryl alcohol-glyoxal and epoxy resins. Int. J. Adhes. Adhes. 94, 47–52.
935 <https://doi.org/10.1016/j.ijadhadh.2019.04.012>
- 936 Zhang, Y., Ding, L., Gu, J., Tan, H., Zhu, L., 2015. Preparation and properties of a starch-based
937 wood adhesive with high bonding strength and water resistance. Carbohydr. Polym.

938 <https://doi.org/10.1016/j.carbpol.2014.08.063>

939 Zhou, L., Han, C., Zhang, S., Gao, Q., Li, H., Li, J., 2012. Preparation and properties of
940 environmental friendly wood adhesives based on wild acorn starch, in: Applied Mechanics
941 and Materials. <https://doi.org/10.4028/www.scientific.net/AMM.121-126.2834>

942 Zhou, X., Du, G., 2020. Applications of Tannin Resin Adhesives in the Wood Industry, in: Tannins
943 - Structural Properties, Biological Properties and Current Knowledge.
944 <https://doi.org/10.5772/intechopen.86424>

945 Zhu, D., Damodaran, S., 2014. Chemical phosphorylation improves the moisture resistance of
946 soy flour-based wood adhesive. J. Appl. Polym. Sci. 131.
947 <https://doi.org/10.1002/app.40451>

948