UNIVERSIDADE FEDERAL DE LAVRAS

DOUGLAS LAMOUNIER FARIA

VALORIZAÇÃO DOS RESÍDUOS AGROINDUSTRIAIS DA CULTURA DE FEIJÃO EM PAINÉIS AGLOMERADOS COM BAIXA EMISSÃO DE FORMALDEÍDO

LAVRAS - MG
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Trabalho de Conclusão de Curso apresentado à Universidade Federal de Lavras, como parte das exigências do Curso de Engenharia de Materiais, para a obtenção do título de Bacharel.

Dr. José Benedito Guimarães Junior
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VALORIZATION OF AGROINDUSTRIAL WASTE FROM BEANS IN PARTICLEBOARDS WITH LOW FORMALDEHYDE EMISSIONS

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APROVADA em 25 de outubro de 2023.

Dr. Thiago de Paula Protásio	UFRA
Dr. Julio Soriano	UNICAMP

Dr. José Benedito Guimarães Junior
Orientador

LAVRAS - MG
2023

A Deus, nossa eterna gratidão.

A meu pai, Vicente, minha mãe Vera, e meu irmão Lucas, que estiveram sempre presentes me apoiando e me dando forças para que eu continuasse na luta durante essa etapa da minha vida. Sempre me senti seguro para continuar.

À minha esposa Daiane, por toda paciência, companheirismo e união em todos os momentos.

Aos amigos e colegas da UEPAM pela amizade.

Ao meu grande amigo Smile (*in memoriam*), por todo companheirismo, lealdade, amor, carinho, alegria, e tudo mais que um filho de quatro patas nos oferece.

AGRADECIMENTOS

À Universidade Federal de Lavras, pela oportunidade concedida.

Ao meu orientador Dr. José Benedito Guimarães Junior por gentilmente ter me ajudado e me guiado durante todos esses anos.

Ao professor Dr. Julio Soriano, sempre prestativo e solícito.

Ao professor Dr. Thiago de Paula Protásio, agradeço pelos ensinamentos práticos com a análise química e escrita acadêmica.

A todos professores que tiveram paciência e sabedoria em transmitir conhecimento ao longo da minha trajetória no curso de Engenharia de Materiais.

A humildade é o primeiro degrau para a sabedoria.

(São Tomás de Aquino)

RESUMO

O formaldeído livre é um carcinógeno cuja redução de emissão em painéis aglomerados vem sendo estudada recentemente para mitigar esse problema ambiental e de saúde humana. Uma alternativa para reduzir a emissão de formaldeído em painéis aglomerados é a utilização de adesivos produzidos a partir de fontes naturais. O cardanol-formaldeído é um adesivo ecológico feito com o líquido da castanha de caju, um subproduto da cadeia do caju. Este trabalho teve como objetivo a produção de painéis aglomerados utilizando cardanol-formaldeído em substituição à ureia-formaldeído. Além disso, diferentes proporções de resíduos de palha de feijão foram utilizadas em substituição à madeira de pinus. A combinação de adesivo ecológico e partículas de resíduos lignocelulósicos pode resultar em um produto que atenda às demandas do mercado e, ao mesmo tempo, não seja agressivo ao meio ambiente. O cardanol-formaldeído promoveu maior ligação interna (0,49 MPa) sobre os painéis colados com ureia-formaldeído, que apresentaram ligação interna de 0,32 MPa. Além disso, o adesivo cardanol-formaldeído promoveu redução de 93% na emissão de formaldeído. Isso significa redução de 16,76 mg/100 g para os aglomerados produzidos com ureia-formaldeído para 1,09 mg/100 g para os produzidos com cardanol-formaldeído. Os resultados indicam o potencial do adesivo à base de cardanol na indústria de painéis de madeira reconstituída.

Palavras-chave: Painel à base de madeira; biomassa residual; materiais lignocelulósicos; adesivo ecológico; poluição ambiental.

ABSTRACT

Free formaldehyde is a carcinogen whose emission reduction in particleboard has been studied recently to mitigate this environmental and human health problem. One alternative to reduce the emission of formaldehyde in particleboards is by using adhesives produced from natural sources. Cardanol-formaldehyde is an environmentally friendly adhesive made with cashew nut liquid, a byproduct from the cashew chain. This work aimed to produce particleboard using cardanol-formaldehyde in place of urea. In addition, different proportions of bean straw wastes were used to replace pine wood. The combination of eco-friendly adhesive and lignocellulosic waste particles could result in a product that meets market demands while being environmentally nonaggressive. Cardanol-formaldehyde promoted greater internal bond (0.49 MPa) on the panels glued with urea-formaldehyde, which presented internal bond of 0.32 MPa. Furthermore, the cardanol-formaldehyde adhesive promoted a 93% reduction in formaldehyde emissions. This means a reduction from 16.76 mg/100 g for particleboards produced with urea-formaldehyde to 1.09 mg/100 g for those produced with cardanol-formaldehyde. The results indicate the potential of cardanol-based adhesive in the reconstituted wood panel industry.

Keywords: Wood-based panel; residual biomass; lignocellulosic materials; eco-friendly adhesive; environment pollution.

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PRIMEIRA PARTE

1 INTRODUÇÃO

A preocupação com produtos ecologicamente corretos e que causam menor agressão ao meio ambiente e à saúde humana tem ganhado atenção nos últimos anos. O bem-estar da sociedade e os anseios por tecnologias mais limpas e com menor potencial danoso ao ser humano são objeto de estudo em diferentes áreas do conhecimento. Dentre estas áreas, a indústria de painéis de madeira reconstituída se destaca com o desafio da produção de painéis com baixa emissão de formaldeído (AKINYEMI, OLAMIDE e OLUWASOGO, 2019). Os painéis de madeira possuem em sua formulação resinas que contém formaldeído e tendem a liberá-lo. Após a reação do formaldeído com a ureia, são formados grupos metilol, estes são instáveis e sofrem hidrólise pela presença da umidade, mesmo à temperatura ambiente. A emissão de formaldeído é uma característica importante ao considerar a compra de painéis de madeira para uso como móveis (YADAV, 2021).

Em 2020 foram produzidos 96 milhões de m³ de painéis aglomerados em todo mundo, um aumento de 24,67% em relação ao volume produzido em 2010 (Food and Agriculture Organization of the United Nations - FAO, 2022). Desse montante, cerca de 95% dos painéis aglomerados são produzidos com adesivos derivados do petróleo contendo formaldeído em sua composição, sendo a ureia-formaldeído o adesivo mais utilizado, em que seu consumo estimado anual é de 11 milhões de toneladas (KUMAR e PIZZI, 2019; PIZZI et al., 2020; HUSSIN et al., 2022). As resinas ureia-formaldeído apresentam um dos mais importantes sistemas de adesivo termorrígido, com o maior consumo de tonelagem na indústria de processamento de madeira devido ao baixo custo, rápida cura, não é inflamável, possui coloração clara e boa solubilidade em água (KELLECI et al., 2022). Porém, possui baixa resistência à água e libera formol (LI et al., 2019).

As preocupações com os efeitos na saúde levaram a uma avaliação de risco mais precisa e à pesquisa e desenvolvimento de novos adesivos com menor impacto na saúde humana e no meio ambiente (BOUSSETTA et al., 2021; CAVALLO et al., 2022; FURTINI et al., 2022), como proteínas, carboidratos, taninos, ligninas e óleos vegetais (DUNKY, 2021). Diversos autores têm estudado o potencial de substituição dos adesivos à base de formaldeído nas indústrias de produção de painéis à base de madeira por outros tipos de adesivos menos tóxicos aos seres humanos e ao meio ambiente (LAMAMING et al., 2020; ISLAM et al., 2020; KARIM et al., 2020; OKTAY, KIZILCAL e BENGU, 2021). Santos et al. (2022) avaliaram o potencial do adesivo à base de extrato de cana-de-açúcar e ácido cítrico para uso em painéis aglomerados. Cavallo et al. (2022) verificaram o potencial de emprego do adesivo orgânico HBP (proteína à base de cânhamo), uma mistura de farinha de cânhamo e reticulador PAE (poliaminoamida epicloridrina) e adesivo inorgânico geopolímero K-PSS (potássio-polisiloxosialato) mais acetato de polivinila em substituição à ureia-formaldeído, sendo verificado emissão de formaldeído e outros COVs (compostos orgânicos voláteis) dez vezes menor que os painéis colados com adesivo comercial ureia-formaldeído. Em trabalho desenvolvido por Furtini et al. (2022), foi utilizado líquido da casca da castanha de caju para produção de painéis aglomerados, sendo verificado melhoria do comportamento à combustão em relação aos painéis aglomerados produzidos com adesivo ureia-formaldeído. Desta forma, alternativas aos adesivos utilizados atualmente para painéis de madeira devem ser considerados, mitigando os efeitos tóxicos e cancerígenos dos adesivos à base de formaldeído.

A necessidade de matérias-primas é outro aspecto inerente à fabricação de painéis aglomerados. Dessa forma, as madeiras de pinus e eucalipto, tipicamente utilizadas para a fabricação de painéis aglomerados, vêm sendo substituídas por biomassa residual da agroindústria (NEITZEL et al., 2022). A produção de aglomerado é um método plausível de converter a biomassa residual em produtos de maior valor e descentralizar a cadeia de matérias-primas (PĘDZIK et al., 2021; GONÇALVES et al., 2022; MARTINS et al., 2021). Essa biomassa residual é gerada após o processamento de culturas agrícolas e florestais para uso na indústria alimentícia e têxtil (NEITZEL et al., 2022), sendo utilizada por diversos pesquisadores na produção de painéis aglomerados como matéria-prima substituta para as tradicionais madeiras de pinus e eucalipto, como resíduo de coco (SOUZA et al., 2022); resíduos de cacau (VELOSO et al., 2020); casca de camélia (CHAYDARREH et al., 2022); sorgo (SUTIAWAN et al., 2022); e resíduos de demolição (AZAMBUJA et al., 2018). Além dessas matérias-primas, os resíduos da cultura do feijão é uma alternativa a ser utilizada na indústria de painéis de madeira. A palha de feijão é uma leguminosa arbustiva do feijoeiro pertencente à família Fabaceae e está presente em diversas regiões do Brasil. O Brasil é o maior produtor mundial de feijão comum (EMPRESA BRASILEIRA DE PESQUISA AGROPECUÁRIA – EMBRAPA, 2022), com área plantada de 2.693,6 mil ha, produtividade de 1.129 kg/ha e produção de 3.040,6 mil t (COMPANHIA NACIONAL DE

ABASTECIMENTO – CONAB, 2023). As biomassas residuais têm várias vantagens, pois são geradas em grandes quantidades, estão disponíveis a um custo consideravelmente baixo, têm baixa densidade e são tipicamente biodegradáveis (FARIA et al., 2021).

Desta forma, este trabalho tem como objetivo avaliar a influência do adesivo cardanol-formaldeído em substituição a ureia-formaldeído e dos resíduos de feijão guandu em substituição à madeira de pinus nas propriedades tecnológicas de painéis aglomerados.

2 OBJETIVOS

2.1 Objetivo geral

Avaliar a influência do adesivo cardanol-formaldeído em substituição a ureia-formaldeído e dos resíduos de feijão guandu em substituição à madeira de pinus nas propriedades tecnológicas de painéis aglomerados.

2.2 Objetivos específicos

- Avaliar a influência dos resíduos de feijão guandu em substituição à madeira de pinus nas resistências mecânica e térmica dos painéis aglomerados.
- Avaliar o teor ideal de resíduos de feijão guandu nos painéis aglomerados.
- Avaliar o efeito do cardanol-formaldeído na entalpia da reação.
- Avaliar o efeito do cardanol-formaldeído na emissão de formaldeído.

3 REFERENCIAL TEÓRICO

3.1 Painéis de madeira

Os painéis de madeira podem ser definidos como produtos compostos de elementos de madeira como lâminas, sarrafos, partículas e fibras, obtidos a partir da redução da madeira sólida e reconstituídas através de ligação adesiva (IWAKIRI, 2020). Desde o início da produção de compensados no final do século XIX, inúmeros tipos de painéis de madeira foram surgindo até o presente momento, sempre com a preocupação de buscar novos produtos com melhor relação custo/benefício, para finalidades específicas a que se destinam.

As chapas de madeira aglomerada surgiram na Alemanha no início da década de 40, como forma de viabilizar a utilização de resíduos de madeira, face a dificuldade de obtenção de madeiras de boa qualidade para produção de painéis compensados, devido ao isolamento da Alemanha durante a 2ª guerra mundial (IWAKIRI, 2020).

As chapas de madeira aglomerada, comercialmente denominado de “aglomerado” no Brasil, é um painel produzido com pequenas partículas de madeira, com a incorporação de um adesivo sintético e reconstituídos numa matriz randômica e consolidados através de aplicação de calor e pressão na prensa quente. Outros materiais lignocelulósicos podem ser utilizados na fabricação de aglomerados (IWAKIRI, 2020).

Entretanto, alguns desafios inerentes às matérias-primas fazem com que os painéis aglomerados apresentem baixa estabilidade dimensional e emitam formaldeído, uma substância comprovadamente cancerígena (IARC, 2023). Devido à natureza hidrofílica das fibras vegetais, os painéis aglomerados quando em contato com umidade tendem a inchar, deixando de desempenhar a função de vedação quando em serviço. Além disso, a origem petrolífera dos adesivos fenólicos utilizados tradicionalmente pelas indústrias de painéis de madeira provoca graves problemas ambientais e de saúde humana, especialmente no tocante à inalação do formaldeído, seja pelos trabalhadores das indústrias de painéis, seja pelo consumidor final.

Os painéis aglomerados são utilizados em diversas aplicações, como na indústria moveleira, que representa 66% de todas as aplicações (EUROPEAN PANEL FEDERATION, 2023), e na construção civil para uso interno, responsável por 27% do total, e utilizado como objetos decorativos e design (FARAG et al., 2020).

3.2 Resíduos agrícolas e agroindustriais

Sabe-se que o Brasil ocupa um lugar de destaque na cadeia produtiva agrícola mundial, e, portanto, a utilização de resíduos desse setor como matéria-prima para painéis pode ser uma alternativa viável, tanto para a destinação adequada dos resíduos quanto para o abastecimento das indústrias de painéis e desenvolvimento de novas tecnologias para o setor. Uma melhor destinação desses resíduos é, sem dúvida, importante, pois promove sua valorização, além de garantir a qualidade sanitária do meio ambiente como um todo (VELOSO et al., 2020).

Biomassas residuais provenientes do agronegócio têm sido utilizadas como substituto às madeiras de pinus e eucalipto, tradicionalmente utilizadas na produção de painéis aglomerados (NEITZEL et al., 2022). As biomassas residuais apresentam diversas vantagens, uma vez que são geradas em grandes quantidades e estão disponíveis a um custo consideravelmente baixo, têm uma baixa densidade e são tipicamente biodegradáveis (CANGUSSU et al., 2021; LAKSONO et al., 2022; AKINYEMI, KOLAJO e ADEDOLU, 2022; SUTIAWAN et al., 2022). Desta forma, o emprego de resíduos lignocelulósicos na produção de painéis aglomerados diversifica os materiais utilizados e agrega valor ao resíduo (PĘDZIK et al., 2021; MARTINS et al., 2021; GONÇALVES et al., 2022). Diante dos inúmeros benefícios observados na literatura quanto ao aproveitamento de resíduos agrícolas, ainda não se sabe os benefícios em termos de redução das emissões de formaldeído (JIMENEZ JUNIOR et al., 2022), mostrando a expectativa de novas contribuições para esta linha de pesquisa.

3.3 Ureia-formaldeído

Esforços têm sido feitos nas últimas décadas para reduzir a liberação de formaldeído. Alguns dos danos que a substância poderia causar à saúde já eram conhecidos há algum tempo. A Agência Internacional de Pesquisa sobre o Câncer (IARC, 2023) citou que o problema ganhou maior visibilidade nos últimos anos, quando o formaldeído foi classificado como cancerígeno. Embora existam alternativas isentas de formaldeído, como poliuretano e resinas acrílicas, que podem ser utilizadas como aglomerantes em compósitos de madeira, as resinas ureia-formaldeído e melamina-formaldeído são amplamente utilizadas por apresentarem as melhores características quanto ao baixo custo e propriedades como não inflamabilidade, taxa de cura rápida e uma cor clara (ROWELL, 2005). A literatura relata um custo de produção de US$ 0,16/kg para ureia. Além disso, o adesivo ureia-formaldeído pode ser produzido a partir da via do óxido metálico (US$ 0,86/kg) e da via da prata (US$ 0,99/kg) (YANG e ROSENTRATER, 2020). Devido ao seu baixo custo, cura rápida, não inflamabilidade, cor clara e boa solubilidade em água, as resinas ureia-formaldeído são um dos sistemas adesivos termoendurecíveis mais importantes na indústria de processamento de madeira, como para produzir painéis de partículas. No entanto, tem baixa resistência à água e libera formaldeído.

Desenvolvida no início da década de 30, a resina ureia-formaldeído possui uma ampla aplicação na indústria madeireira em todo o mundo, na colagem de madeira sólida, compostos laminados e particulados em geral. Mais de 90% dos produtos compostos de madeira utilizam esta resina, tendo em vista o seu baixo custo em relação às outras resinas (IWAKIRI, 2020).

No entanto, uma das desvantagens, consiste na sua susceptibilidade a degradação hidrolítica na presença de umidade e/ou ácidos, especialmente em temperaturas moderadas a elevadas. Enquanto que a quebra da estrutura da resina é muito lenta em água fria, a deterioração se acelera acima de 40 ºC e torna-se muito rápida a temperatura acima de 60 °C. Portanto, esta resina é classificada como de uso interno (IWAKIRI, 2020).

3.4 Líquido da casca da castanha de caju

O líquido da casca da castanha de caju (CNSL) é um subproduto da indústria do caju, plantas da família das Anacardiaceae, como o cajueiro (*Anacardium occidentale* Linn), um subproduto agrícola amplamente disponível no Nordeste Brasileiro. O CNSL representa aproximadamente 25% do peso da castanha e é considerado um subproduto de agronegócio do caju, de baixíssimo valor agregado. Possui composição química composta por hidrocarbonetos fenólicos de cadeia longa que depois de extraído, apresenta coloração amarelo-esverdeado e alta viscosidade (SUMMERTON, HURST e CLARK, 2018). Dentre os compostos presentes no CNSL, o cardanol se destaca por possuir alto isolamento elétrico e boa estabilidade térmica. Sua estrutura possui ampla funcionalidade com três sítios reativos, hidroxila fenólica, anel aromático e insaturação na cadeia lateral alquenila (JADHAV et al., 2018). O CNSL é composto de ácido anacárdico (74-77%), cardanol (1-9%), cardol (15-20%) e 2-metil cardol (1-3%) (CAILLOL, 2018). O cardanol é extraído ou refinado do CNSL após processamento a quente e extraído por calor durante a torrefação da noz para separar a amêndoa, onde o ácido anacárdico é descarboxilado em cardanol após o aquecimento. O cardanol é um composto fenólico com uma cadeia carbonada alifática C15 substituída na posição meta que apresenta insaturação e saturação (CITÓ et al., 1998). Pode ser polimerizado por diversos caminhos, sendo os mais comuns a produção de resinas alquídicas através da polimerização por adição na cadeia lateral e a produção de resinas fenólicas através da polimerização por condensação com aldeídos (FRANÇA, 2007). Na polimerização por condensação com formaldeído, o cardol apresenta maior reatividade devido à presença de duas hidroxilas no anel aromático. Este fato favorece a polimerização seletiva dos monômeros fenólicos do CNSL (FRANÇA, 2007). O cardanol parece ser um substituto sustentável para adesivos derivados de petróleo convencionais porque é de origem vegetal, tem baixa volatilização e não é tóxico (UDHAYASANKARA et al., 2018).

Tomkinson (2002) utilizou o líquido da casca da castanha de caju (CNSL) reduzido por ozonólise para fazer painéis aglomerados e demonstrou uma melhora na adesão entre as partículas e o adesivo aldeído CNSL quando comparado ao fenol-formaldeído. Assim, alternativas aos adesivos atualmente utilizados para painéis de madeira devem ser consideradas, mitigando os efeitos tóxicos e cancerígenos dos adesivos à base de formaldeído. No entanto, a reposição do formaldeído é dificultada por questões técnicas e econômicas. Isso implica que o desenvolvimento de um adesivo alternativo e menos nocivo envolve a compatibilidade química do novo componente a ser testado e seu custo, conforme mencionado acima, já que a ureia-formaldeído é reconhecidamente vantajosa economicamente.

Diversas aplicações tecnológicas têm utilizado o cardanol, como em matrizes termofixas em compósitos reforçados com fibras de buriti (SANTOS et al., 2010), bagaço de cana-de-açúcar (BALAJI et al., 2018), madeira laminada densificada (SHISHLOV et al., 2015), polímeros e aditivos de base biológica (CAILLOL, 2018) e adesivos para painéis de partículas (FURTINI et al., 2022).

4 CONSIDERAÇÕES FINAIS SOBRE A REVISÃO BIBLIOGRÁFICA

Diante do que foi apresentado nos tópicos anteriores, a reciclagem de resíduos provenientes da agroindústria é uma opção para mitigar os impactos ambientais causados pelo descarte incorreto destes materiais. A valorização de resíduos em produtos de maior valor agregado se mostra uma alternativa ecologicamente, socialmente e financeiramente viável, contribuindo para inserir as indústrias brasileiras no contexto da “economia circular” e “economia verde”. Os painéis aglomerados são materiais já estabilizados nas indústrias moveleiras e da construção civil. A produção de painéis aglomerados utilizando resíduos da cultura de feijão se mostra uma alternativa às madeiras de eucalipto e pinus, tradicionalmente utilizadas pelas indústrias produtoras de painéis reconstituídos, reduzindo a disposição irregular dos resíduos provenientes da cultura de feijão, além de contribuir para a diversificação das matérias-primas empregadas na cadeia produtiva de painéis.

Além disso, a substituição da ureia-formaldeído pelo cardanol-formaldeído, um adesivo ecologicamente correto na produção de painéis aglomerados é uma proposta com possíveis aplicações práticas nas indústrias moveleiras e da construção civil. Por ser de origem vegetal, não tóxico e por emitir baixa volatização, se torna uma alternativa sustentável na substituição de substâncias agressoras ao meio ambiente e à saúde humana, como é o caso dos painéis aglomerados produzidos com adesivo à base de formaldeído.

Entretanto, a literatura carece de estudos envolvendo a produção de painéis aglomerados utilizando resíduos da cultura de feijão, bem como o comportamento dos painéis empregando cardanol-formaldeído.

Desta forma, este trabalho foi desenvolvido em um artigo visando à caracterização química e térmica do cardanol-formaldeído, bem como a influência de diferentes teores de substituição da madeira de pinus pelos resíduos de feijão, a fim de se entender as propriedades tecnológicas dos painéis aglomerados.

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SEGUNDA PARTE - ARTIGO

Publicado em 09/02/20223 no periódico Environmental Science and Pollution Research

(Impact factor 5.8)

Cardanol-based adhesive with reduced formaldehyde emission to produce particleboards with waste from bean crops

https://link.springer.com/article/10.1007/s11356-023-25764-5

Douglas Lamounier Faria[1*]; Mário Vanoli Scatolino[2]; Juliano Elvis de Oliveira[3];
Fabricio Gomes Gonçalves[4]; Julio Soriano[5]; Thiago de Paula Protásio[6];
Roberto Carlos Costa Lelis[7]; Luisa Maria Hora de Carvalho[8];
Lourival Marin Mendes[1]; José Benedito Guimarães Junior[3]

[1]Department of Forest Science, Federal University of Lavras - UFLA, Perimetral Av., POB 3037, 37200-900, Lavras, MG, Brazil. Tel: +5535999377919; E-mail: douglas.lamounier@yahoo.com (*corresponding author); lourival@ufla.br

[2]Agricultural Sciences Center, Federal University of the Semiarid - UFERSA, 59625-900, Mossoró, RN, Brazil. Email: marioscatolino@gmail.com

[3]Department of Engineering, Federal University of Lavras - UFLA, Perimetral Av., POB 3037, 37200-900, Lavras, MG, Brazil. E-mail: juliano.oliveira@ufla.br, jose.guimaraes@ufla.br

[4]Department of Forest and Wood Sciences, Federal University of Espírito Santo - UFES, Governador Carlos Lindemberg Av., 316, 29550-000, Jerônimo Monteiro, ES, Brazil. E-mail: fabricio.goncalves@ufes.br

[5]School of Agricultural Engineering – University of Campinas – UNICAMP, Candido Rondon Av, 13083-875, Campinas, SP, Brazil. E-mail: julio.soriano@feagri.unicamp.br

[6]Federal Rural University of the Amazon - UFRA, Parauapebas, PA, Brazil. E-mail: thiago.protasio@ufra.edu.br

[7]Department of Forest Products, Forest Institute, Federal Rural University of Rio de Janeiro - UFRRJ, BR-465, km 07, 23890-000, Seropédica, RJ, Brazil. E-mail: lelis@ufrrj.br

[8] Department of Wood Engineering, School of Technology and Management of Viseu – ESTGV, Campus Politécnico, 3504-510 Viseu, Portugal. E-mail: lhcarvalho@estgv.ipv.pt

ORCID:
Douglas Lamounier Faria: (https://orcid.org/0000-0002-5405-8430)
Mário Vanoli Scatolino: (https://orcid.org/0000-0002-0412-994X)
Juliano Elvis de Oliveira: (https://orcid.org/0000-0001-9570-8308)
Fabricio Gomes Gonçalves: (https://orcid.org//0000-0003-2010-9508)
Julio Soriano: (https://orcid.org/0000-0002-3446-532X)
Thiago de Paula Protásio: (https://orcid.org/0000-0002-5560-8350)
Roberto Carlos Costa Lelis: (https://orcid.org/0000-0003-2923-3839)
Luisa Maria Hora de Carvalho: (https://orcid.org/0000-0003-4557-7113)
Lourival Marin Mendes: (https://orcid.org/0000-0001-8713-405X)
José Benedito Guimarães Junior: (https://orcid.org/0000-0002-9066-1069)

Abstract

Free formaldehyde is a carcinogen whose emission reduction in particleboard has been studied recently to mitigate this environmental and human health problem. One alternative to reduce the emission of formaldehyde in particleboards is by using adhesives produced from natural sources. Cardanol-formaldehyde is an environmentally friendly adhesive made with cashew nut liquid, a byproduct from the cashew chain. This work aimed to produce particleboard using cardanol-formaldehyde in place of urea. In addition, different proportions of bean straw wastes were used to replace pine wood. The combination of eco-friendly adhesive and lignocellulosic waste particles could result in a product that meets market demands while being environmentally nonaggressive. Cardanol-formaldehyde promoted a higher modulus of elasticity (MOE) (1172 MPa) and modulus of rupture (MOR) (4.39 MPa) about panels glued with urea-formaldehyde, which presented a MOE of 764 MPa and MOR of 2.45 MPa. Furthermore, the cardanol-formaldehyde adhesive promoted a 93% reduction in formaldehyde emission, with a reduction from 16.76 mg/100 g to 1.09 mg/100 g oven-dry board for particleboards produced with cardanol-formaldehyde, indicating potential as an adhesive in the particleboard industry.

Keywords: Wood-based panel; waste; lignocellulosic materials; eco-friendly adhesive; low-emission formaldehyde; physical and mechanical properties.

1. Introduction

The concern with ecologically correct products that cause lower environmental and human health aggression has gained attention in recent years. The well-being of society and the desire for cleaner technologies with lower harmful potential for human beings are the objects of study in different areas of knowledge. Among these areas, the reconstituted wood-based industry stands out with the challenge of reducing formaldehyde emissions in particleboard production. Wood-based panels contain resins with formaldehyde in their composition and tend to release it. After the reaction of formaldehyde with urea, methylol groups are formed. These are unstable and undergo hydrolysis by moisture, even at room temperature. Formaldehyde emissions are an important feature when considering the purchase of wood-based panels for use as furniture (Yadav 2021).

Efforts have been made over the last few decades to reduce formaldehyde release. Some of the damage the substance could present to health was already known for some time. The International Agency for Research on Cancer (IARC 2016) cited that the problem gained greater visibility in recent years when formaldehyde was classified as a carcinogen. Although there are formaldehyde-free alternatives, such as polyurethane and acrylic resins, which can be used as binders in wood composites, urea-formaldehyde and melamine-formaldehyde resins are widely used because they have the best characteristics regarding low cost and properties such as nonflammability, rapid cure rate, and a light color (Rowell 2005). The literature reports a production cost of \$0.16/kg for urea. Additionally, urea-formaldehyde adhesive can be produced from the metal oxide pathway (\$0.86/kg) and the silver pathway (\$0.99/kg) (Yang and Rosentrater 2020). Because of their low cost, fast curing, nonflammability, light color, and good water solubility, urea-formaldehyde resins are one of the most important thermosetting adhesive systems in the wood processing industry (Dunky 1998), such as to produce particleboards. However, it has low water resistance and releases formaldehyde (Popovic et al. 2011). Particleboards are used in a range of applications, such as in the furniture industry, which represents 66% of all applications (European Panel Federation, 2023), and in civil construction for use indoors, responsible for 27% of the total, and used as decorative objects and design (Farag et al. 2020). In 2020, 96 million m^3 of particleboard were produced worldwide, an increase of 24.67% compared to the volume produced in 2010 (Food and Agriculture Organization of the United Nations - FAO 2022). Approximately 95% of particleboards are made with adhesives derived from petroleum containing formaldehyde in their composition, with urea-formaldehyde being the most commonly used adhesive, with an estimated annual consumption of 11 million t (Kumar and Pizzi 2019; Pizzi et al. 2020).

Concerns about health effects have led to a more accurate risk assessment and to the development of new adhesives that have a lower impact on human health and the environment (Cavallo et al. 2022), such as proteins, carbohydrates, tannins, lignins, and vegetal oils (Dunky 2021). Several authors have studied the potential for replacing formaldehyde-based adhesives in wood-based panel production industries with other less toxic adhesive types for humans and the environment. Santos et al. (2022) assessed the adhesive potential for use in particleboards based on sugarcane extracts and citric acid. When compared to commercial adhesives, particleboard made with organic adhesive derived from hemp protein had a significantly lower level of free formaldehyde (Cavallo et al. 2022). Tomkinson (2002) used ozonolysis-reduced cashew nut shell liquid (CNSL) to make particleboards, and an improvement in adhesion between the particles and the CNSL aldehyde adhesive was demonstrated when compared to phenol-formaldehyde. Thus, alternatives to the adhesives currently used for wood panels should be considered, mitigating the toxic and carcinogenic effects of formaldehyde-based adhesives. However, formaldehyde replacement is hampered by technical and economic issues. This implies that the development of an alternative and less harmful adhesive involves compatibility chemistry on the new component to be tested and its cost, as mentioned above since urea-formaldehyde is known to be economically advantageous. CNSL is produced as a byproduct of cashew processing, corresponding to approximately 25% of the nut mass; plants of the Anacardiaceae family, such as cashew (*Anacardium occidentale* Linn), an agricultural byproduct widely available in Northeast Brazil (Ionescu et al. 2012). It has a chemical composition composed of long-chain phenolic hydrocarbons that, after being extracted, have a greenish-yellow color and high viscosity (Summerton et al. 2018). CNSL is composed of anacardic acid (74-77%), cardanol (1-9%), cardol (15-20%), and 2-methyl cardol (1-3%) (Caillol 2018). Among the compounds present in CNSL, cardanol stands out as having high electrical insulation and good thermal stability. Its structure has broad functionality with three reactive sites, phenolic hydroxyl, aromatic ring, and unsaturation in the alkenyl side chain (Jadhav et al. 2018). Cardanol is extracted or refined from the CNSL after hot processing and heat extracted during nut roasting to separate the kernel, where the anacardic acid decarboxylates to cardanol after heating (Lochab et al.

2014). Cardanol is a phenolic compound with a C15 aliphatic carbon chain substituted in the meta position that presents unsaturation and saturation (Citó et al. 1998). It can be polymerized in a variety of ways, the most common being the production of alkyd resins via side-chain addition polymerization and the production of phenolic resins via condensation polymerization with aldehydes (Moita Neto 1997; França 2007). In polymerization with formaldehyde, cardanol shows greater reactivity because of the presence of two hydroxyl groups in the aromatic ring. This favors the selective polymerization of CNSL phenolic monomers. Urea-formaldehyde resin for particleboard at the end of the 1970s would have had a formaldehyde/urea molar ratio of approximately 1.6 - 1.8. Today, urea-formaldehyde for the same application has a reduced molar ratio of between 1.02 and 1.08 (Dunky and Pizzi 2002). Cardanol-formaldehyde resins are prepared in a range of 0.6 – 0.9 of the formaldehyde/cardanol ratio (Bajpai et al. 2008). Dazmiri et al. (2019) explained that lower formaldehyde/urea molar ratios showed relatively poor adhesion when used to manufacture wood-based composites.

Cardanol seems to be a sustainable substitute for conventional petroleum-derived adhesives because it comes from a vegetable source, has low volatilization and is nontoxic (Udhayasankar et al. 2018). Several technological applications have used cardanol, such as in thermoset matrices in composites reinforced with buriti fibers (Santos et al. 2010), sugarcane bagasse (Balaji et al. 2017; Balaji et al. 2018), densified laminated wood (Shishlov et al. 2015), biobased polymers and additives (Caillol 2018), and adhesives for particleboards (Furtini et al. 2022).

The necessity for raw materials is another inherent aspect of particleboard manufacturing. In this way, pine and eucalyptus wood, which are typically used to make particleboard, have been replaced with residual biomass from the agricultural industry (Neitzel et al. 2022). Particleboard production is a plausible method of converting remaining biomass into higher-value products and decentralizing the raw materials chain (Pędzik et al. 2021; Gonçalves et al. 2022; Martins et al. 2021). Such residual biomass is generated after the processing of agricultural and forestry crops for use in food and the textile industry (Neitzel et al. 2022), being used by several researchers in the production of particleboards as substitute raw materials for traditional pine and eucalyptus wood, such as coconut waste (Souza et al. 2022); cocoa waste (Veloso et al. 2020a); tea oil camellia shell (Chaydarreh et al. 2022); sorghum (Sutiawan et al. 2022); residues of CCB-treated pinus (Bertolini et al. 2013); and demolition wastes (Azambuja et al. 2018). In addition to this raw material, bean crop wastes are an alternative to be used in the wood panel industry. Bean straw is a bean shrub legume that belongs to the Fabaceae family and is present in different parts of Brazil. Despite not being one of the most popular beans in the country, it has great importance in the agro-industrial chain. Brazil is the world's largest producer of common bean (Empresa Brasileira de Pesquisa Agropecuária - Embrapa 2022), with a planted area of 2,816.1 thousand ha, a productivity of 1095 kg/ha, and a production of 3,083.6 thousand t (Companhia Nacional de Abastecimento – Conab 2022). Residual biomasses have several advantages since they are generated in large quantities, are available at a considerably low cost, have a low density and are typically biodegradable (Ku et al. 2011; Faruk et al. 2012). In addition, the use of these residues in particleboards reduces the deforestation of planted forests and promotes their correct destination, reducing the emission of greenhouse gases since such residues are generally burned or disposed of irregularly in landfills or wasteland (Drovou et al. 2022; Quereshi et al. 2022). Given the numerous benefits seen in the literature regarding the use of agricultural residues, the benefits in terms of reducing formaldehyde emissions are still unknown (Jimenez Jr et al. 2022), showing the expectation for new contributions for this line of research.

Thus, to provide wood-based products with increasing environmental benefits in particleboard, this research aimed to evaluate the benefits obtained by substituting the urea-formaldehyde adhesive with cardanol-formaldehyde in terms of formaldehyde emission. The technological properties of particleboards produced with different contents of residual straw of the bean crop, in partial substitution of pine wood particles, were evaluated.

2. Material and methods

2.1 Procurement and characterization of raw materials

The particleboards were made from bean waste (*Cajanus cajan* L. Mill sp) and wood from 28-year-old pine (*Pinus oocarpa*). The wood was collected on the campus of the Federal University of Lavras (UFLA), located in the municipality of Lavras in the southern region of Minas Gerais, Brazil; coordinates 21° 14′ 45″ S, 44° 59′ 59″ W, and the altitude 920 m. Two trees were harvested and logs with 0.60 m from the bases were crushed in a hammer mill. Bean wastes were obtained from the Experimental Farm Vitorinha in the municipality of Ijaci, Minas Gerais (21° 10’ S, and 44° 55’ W, altitude 951 m). They were processed in a hammer mill to obtain sliver particles. To reduce the moisture content of the pine wood and bean waste particles to a final value of 3%, they were oven-dried at 105 ± 2 °C for 24 h. The particles selected to make the particleboards ranged in granulometry from 10 mesh (2.00 mm) to 30 mesh (0.595 mm).

The basic density of bean particles was obtained after particle saturation and subsequent volume measurement in the measuring cylinder. After drying the particles (105 ± 2 °C for 24 h), the basic density was obtained by the ratio between the dry mass/saturated volume of the particles, as described in Scatolino et al. (2019). The basic density of the pine wood was determined by following the guidelines of the NBR 11941 (Brazilian Association of Technical Standards - ABNT 2003) standard, in which discs were obtained from each tree at a height of 1.30 m from the ground, and the discs were cut into four wedges. Two wedges were used to determine the basic density, and two wedges were used for chemical characterization.

After grinding the raw materials in a knife mill, the chemical constituents were determined using the fraction that passed through 40 mesh (0.420 mm) and was retained on 60 mesh (0.250 mm). The following standards were used for determining the chemical constituents of lignocellulosic materials: total extractive content NBR 14853 (ABNT 2010); insoluble lignin content NBR 7989 (ABNT 2010), and ash content NBR 13999 (ABNT 2017). The holocellulose content was obtained based on the procedures described by Browning (1964).

2.2 Synthesis and characterization of cardanol-formaldehyde adhesive

The cardanol-formaldehyde adhesive was synthesized similarly to the work of Santos et al. (2010). A 2.0 M NaOH solution was used as a catalyst in the polycondensation reaction using a ratio of 1:5 formaldehyde to cardanol (Resibras Cashol). First, cardanol was mixed with formaldehyde under constant stirring in a water bath at 90 °C for 60 min to homogenize the reagents. Subsequently, as it is a polycondensation reaction, the catalyst was dripped over 60 min to avoid a rapid increase in the polymeric mass and to control the viscosity (Figure 1).

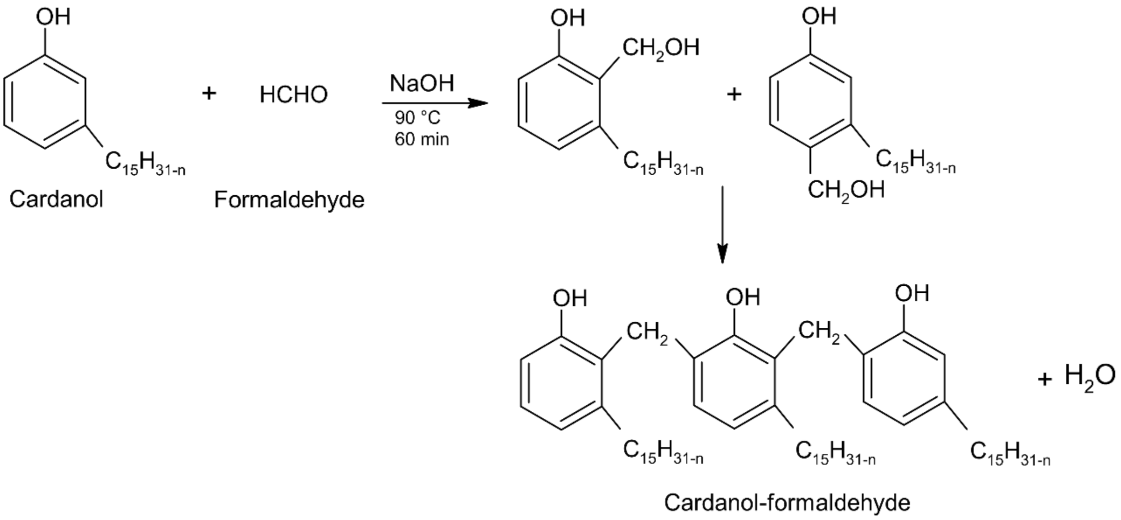

Fig. 1 Cardanol-formaldehyde reaction

Differential scanning calorimetry (DSC) was performed to verify the adhesive behavior during pressing using Q100 TA Instruments in a nitrogen atmosphere with a flow rate of 50 mL/min. Five milligrams of adhesive were placed in sealed pans, and the test was performed at a temperature of 25 °C for 20 min, then increased at a rate of 50 °C/min to a temperature of 160 °C, where it remained for 2 h at a rate of 10 °C/min. By integrating the reaction peak area and the baseline interpolated between its start and finish, the polymerization reaction enthalpy was calculated. Infrared spectra were obtained using a Nicolet 470 Nexus FTIR spectrometer operating in transmission mode. The FTIR spectrometer was continuously purged throughout the nitrogen analysis. A total of 64 scans were collected with a resolution of 2 cm^{-1} for each spectrum (4000-400 cm^{-1}).

2.3 Production of the particleboards

Particleboards with dimensions of 250 x 250 x 15 mm (length, width, and thickness, respectively) and a nominal density of 0.64 g/cm^3 were manufactured. Four types of particleboards were made by replacing different levels of pine with bean waste using cardanol-formaldehyde. For comparison and validation of the proposed substitution of particles, pine particleboards (named Control) were produced containing urea-formaldehyde adhesive in their composition (Table 1). Each composition consisted of three replications, totalling 15 boards.

Table 1 Compositions of particleboards

Composition	Pine (%)	Bean waste (%)	Adhesive formulation
Control	100	0	Urea-formaldehyde
0% bean	100	0	Cardanol-formaldehyde
5% bean	95	5	
10% bean	90	10	
15% bean	85	15	

The pine and bean waste were placed in a rotating drum and sprayed with 8% cardanol-formaldehyde adhesive (solids content of 73.57%, pH of 6.23, and viscosity of 1.55 Pa·s) for 8 min. The control particleboard was produced with urea-formaldehyde (solids content of 70.0%, pH of 8.45, and viscosity of 1.81 Pa·s). Adhesives with extremely low pH can degrade lignocellulosic particles, whereas adhesives with extremely high pH can cause foaming at the glue line. In addition, pH directly affects protonation and deprotonation, influencing adhesive polymerization. The pH range used in this work did not cause any problems.

The mixture of particles and adhesive was placed in a metallic box mattress forming (300 × 300 × 15 mm). The mixture was prepressed at 0.5 MPa for 5 min at room temperature to form the “mattress”. Afterward, the mixture of particles and urea-formaldehyde adhesive were pressed at 160 °C at a specific pressure of 4.0 MPa for 8 min, whereas the composition of particles with cardanol-formaldehyde was pressed at 160 °C at a specific pressure of 4 MPa for 13 min (Figure 2). The formed particleboards were kept in a climatic chamber (20 ± 3 °C; relative humidity 65 ± 5%) until equilibrium moisture (~12%) and complete adhesive polymerization were achieved.

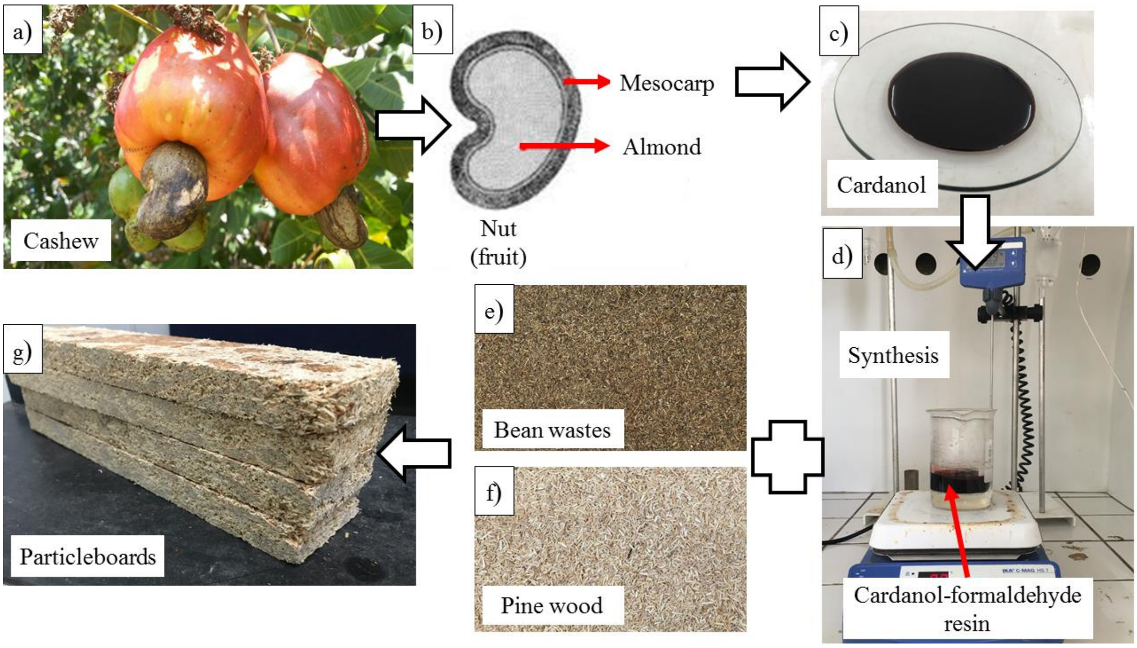

Fig. 2 Production of the particleboards; a) cashew, adapted from Pixabay (2022); b) nut (Agência UFC 2022); c) cardanol-formaldehyde adhesive; d) synthesis of cardanol-formaldehyde; e) bean wastes; f) pine wood; g) samples of the particleboards

2.4 Physical, mechanical, and combustion properties of particleboards

The particleboard moisture content (basis dry basis) was obtained using five specimens with dimensions of 50 × 50 mm according to NBR 14810–2 (ABNT 2018). The bulk density was evaluated using five specimens in 50 × 50 mm dimensions following the procedures described in D1037-12 (American Society for Testing and Materials - ASTM 2020).

According to Scatolino et al. (2017), the compression ratio was calculated by dividing the bulk density of the test specimens by the basic density of the pine and bean particles. To obtain the thickness swelling after 2 and 24 h immersion in water (TS 2 h and TS 24 h), five specimens with dimensions of 50 × 50 mm were cut from each particleboard. The tests were performed according to the standard procedures D1037-12 (ASTM 2020).

The vertical density profile of the samples (50 × 50 × 15 mm) was determined by an X-ray densitometer in a range of 20 μm and a density profile meter (Grecon, DAX-6000, Germany). Five samples were measured for each panel type.

Five specimens with dimensions of 50 x 50 x 15 mm were used to measure the internal bond (IB) by the D1037-12 standard (ASTM 2020). The modulus of rupture (MOR) and modulus of elasticity (MOE) were determined by the three-point static bending test based on the D1037-12 standard (ASTM 2020). Three specimens of each composition with 250 mm in length, 50 mm in width, and 15 mm in thickness were used, with the supports positioned at a span of 200 mm. The specimens were tested in an Arotec servo-electric universal press testing machine equipped with a 20 kN load cell with a 5 mm/min test speed. Figure 3 shows the performance of the static bending and internal bond tests.

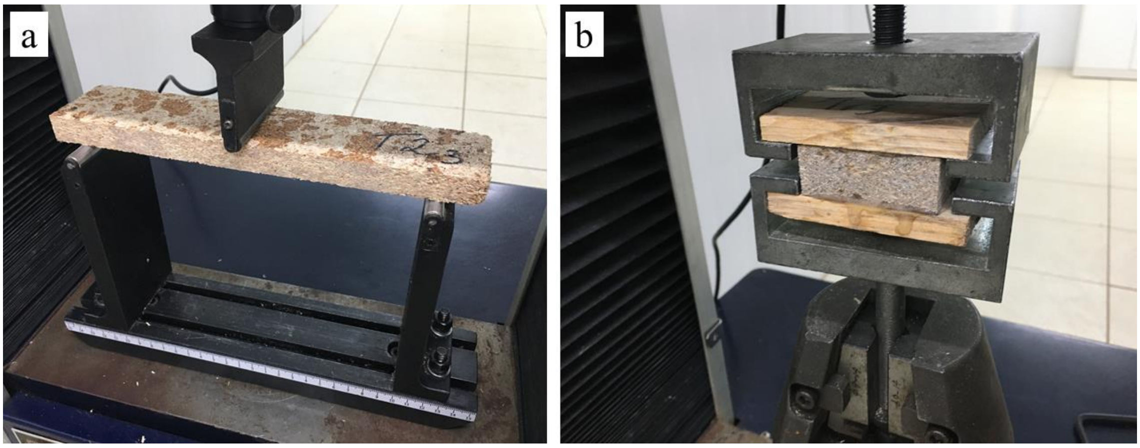

Fig. 3 Static bending and internal bond tests in particleboards

The particleboard combustibility was determined according to Quirino and Brito (1991) and Paula et al. (2011). The test was carried out in a combustor made of a galvanized iron sheet containing an aluminum and wood base, a temperature controller, a scale with a precision of 0.05 g, and an aluminum shield surrounding the combustor to avoid wind interference during combustion. For each composition studied, three specimens with dimensions of 50 × 50 × 15 mm were used and later sectioned into four samples of 25 × 25 × 15 mm. The base and the combustor were placed on a balance. The specimens were inserted, and their mass was measured. Then, the screen and the temperature gauge were placed. The ignition started with the combustion of 20 mL of alcohol deposited on an aluminum plate located on the metal base and below the combustor.

2.5 Formaldehyde emission of the particleboards

The flask method is simple; nevertheless, it meets today's requirements regarding quantitative analysis, such as high reproducibility and the capacity to detect small differences. In this work, the free formaldehyde emission of particleboard was tested according to the standard EN 717-3 (EUROPEAN STANDARD - EN 1996) using the flask method.

The prepared test samples were suspended with a special device over 50 mL distilled water in a 500 mL capacity polyethylene flask. The sealed flasks were left for 3 h in a drying oven at 40 °C. These were then cooled to room temperature to achieve complete absorption of the formaldehyde in water. Quantification was performed by UV/VIS spectroscopy at 412 nm using the acetylacetone method.

2.6 Surface morphology

The particleboards were analyzed by scanning electron microscopy (SEM). For this, the specimens were covered with a layer of gold in an evaporator and analyzed in a LEO EVO 40 XPV scanning electron microscope operated at 20 kV.

2.7 Statistical analysis

The data were analyzed by considering completely randomized designs to assess the particleboard properties. The findings were submitted to analysis of variance (ANOVA), with least significant differences (LSD) and regression at a 5% significance level, to assess the inclusion of particles of bean wastes in the composition of particleboards. Data were processed using Sisvar 5.6 software.

3. Results and discussion

3.1 Characterization of the cardanol-formaldehyde adhesive

The DSC thermograms identified a single exothermic peak at 33 min (Figure 4). This thermal event was significant for understanding adhesive polymerization during pressing. As observed in the isotherm, the peak verified at 33 min showed that, as the analysis remained at 25 °C for 20 min, the thermal event seen at the pressing temperature of 160 °C occurred at 13 min. This point can be associated with the degradation of organic matter (Hýsek and Zółłtowska 2022) and with adhesive crosslinking (Oktay et al. 2021). The polymerization process occurs through the formation of a chain of cross-links between the hydroxyls of the cardanol-formaldehyde adhesive, resulting in an enthalpy of -299.41 J/g. According to Xing et al. (2005), the curing enthalpy of the UF resin decreased with the presence of wood materials. However, the decrease in resin enthalpy of curing is more affected by the presence of bark particles than by wood.

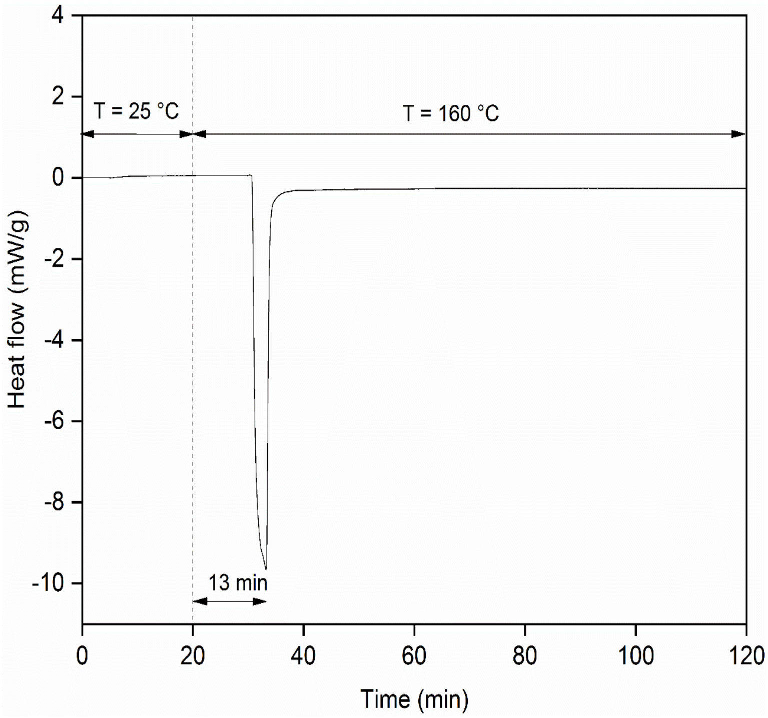

Fig. 4 DSC isotherm for cardanol-formaldehyde adhesive

The FTIR spectrum observed was similar to that obtained for the phenol-formaldehyde adhesive. This result was expected since cardanol is a phenolic compound (Figure 5). Furthermore, it was possible to verify that the polymerization occurred by the aromatic ring and not by the double bonds of the aliphatic chain due to the presence of the peak at 690 cm^{-1}, related to the double bonds of the C=C side chain (Mwaikambo and Ansell 2001).

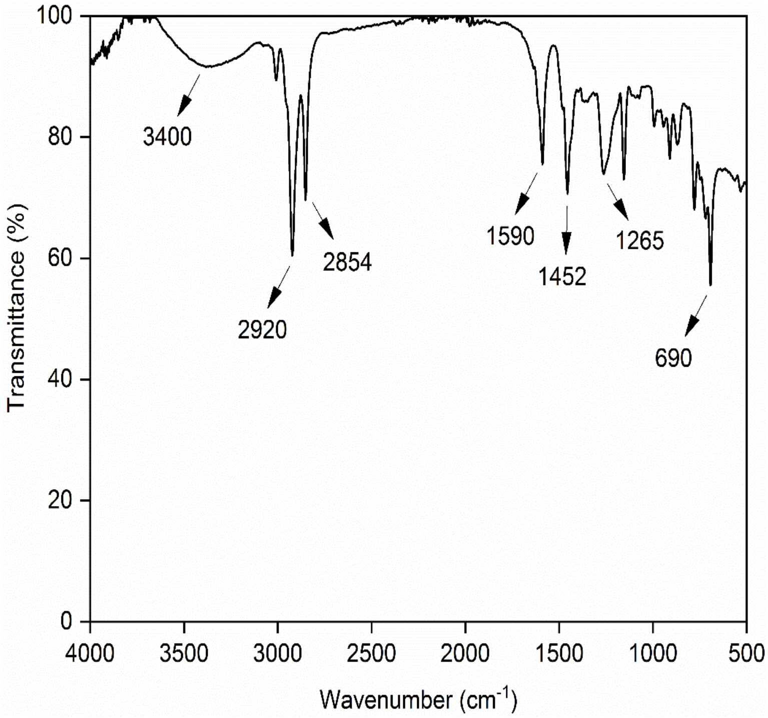

Fig 5 FTIR spectra of cardanol-formaldehyde adhesive

The broad peak of phenolic hydroxyl groups at 3400 cm^{-1} corresponds to the axial deformation of the hydroxyl groups present in the adhesive (Shrestha et al. 2018). This band is extensive because this group forms van der Waals bonds (hydrogen bonds). At 2920 cm^{-1} there is a strong peak confirming the presence of axial deformation of the CH_2 group in cardanol, while at 2854 cm^{-1} is the –CH stretching aliphatic linkage (Santos et al. 2021). The peak at 1590 cm^{-1} is because of the skeletal vibrations of the aromatic C=C linkages (Cong et al. 2014), and the peak observed at 1452 cm^{-1} corresponds to the axial deformation of the C=C double bonds in the aromatic ring (Zhang et al. 2021). The peak at 1265 cm^{-1} corresponds to the presence of a C-O stretching aromatic ring, whereas the peak at 690 cm^{-1} is attributed to the specific absorptions of aromatic rings (aromatic CH bending) (Yusof et al. 2013). Similar results were reported (Mwaikambo and Ansell 2001; Balaji et al. 2017).

3.2 Physical and chemical characterization of pine wood and bean waste

The pine particles presented a basic density of 0.477 g/cm³, while the bean straw particles showed a value of 0.123 g/cm³. One of the main factors inherent to the quality of the boards produced is the basic density (Iwakiri 2005). Due to the low basic density of bean waste particles, more particles are required to obtain the nominal density, which directly affects the compression ratio (Martins et al. 2021). According to Maloney (1993), species with a density of up to 0.550 g/cm³ are the most suitable because the compression ratio ranges from 1.3 to 1.6. In this way, replacing particles of lower basic density will promote a higher compression ratio for the boards produced, resulting in improved mechanical properties due to the reduction of voids. However, the greater number of particles with low basic density reduces the dimensional stability due to the greater number of hydroxylic sites (Kelly 1977). In addition to the basic density, other essential variables, such as chemical composition, production process, and type of adhesive, must be considered (Maloney 1993; Tsoumis 1991; Guimarães Junior et al. 2016). The waste straw from bean crops presented a higher content of total extractives than the pine wood (Table 2).

Table 2 Chemical composition of pine wood and bean wastes

Chemical component	Pine wood (%)	Bean wastes (%)
Total extractives[1]	4.03 ± 0.17*	21.62 ± 2.15*
Insoluble lignin[2]	26.08 ± 2.50*	29.72 ± 2.95*
Holocellulose[2]	69.63 ± 6.08*	46.80 ± 5.64*
Ash[1]	0.26 ± 0.02*	1.86 ± 0.08*

*Standard deviation; [1]dry basis; [2]extractives-free dry basis.

Extractives are secondary metabolites with low molecular weights produced by plants with the function of attraction or protection, generally terpenes, waxes, oils, greases, resins, alkaloids, and tannins (Fengel and Wegener 1983). High contents of total extractives block the contact between the adhesive and the particle, promoting poor adhesion and decreasing the mechanical properties (Bufalino et al. 2012). It has been reported in the literature that nonwood lignocellulosic materials have higher extractive contents, as verified by Faria et al. (2022). Their authors obtained total extractive contents of 3.6% for *Eucalyptus* spp. and 12.4% for *Mauritia flexuosa* wastes. The same was observed by Veloso et al. (2020a), who obtained total extractives of 4.0% and 34.8% for pine wood and cocoa wastes, respectively.

For the lignin content, similar contents resulted in pine wood and bean wastes (Table 2). Lignin is a natural adhesive formed by phenylpropane units, presenting a highly condensed structure and irregular appearance that provides high rigidity for the material. It acts as a damper for the cellulose microfibrils, limiting the movement parallel to the grain and increasing the mechanical strength to external forces (Sweet and Winandy 1999). Higher contents of this component are desired for producing higher-quality particleboards. Lignin promotes the cohesiveness of particles, which consequently improves the adhesion of particleboards and increases the board's mechanical properties.

Bean wastes had lower holocellulose content compared to pine wood. Holocellulose refers to the sum of cellulose and hemicellulose values. Therefore, increased holocellulose contents tend to reduce the dimensional stability of the particleboard since the latter component is the most hygroscopic component of the plant cell wall (Iwakiri 2005). Thus, the lowest contents of holocellulose in bean wastes are interesting because of the lower hygroscopicity, and the panels result in superior physical properties.

The ash contents in bean wastes were seven times higher than those found in pine wood (Table 2). Higher ash concentrations can alter the pH, affecting adhesion and mechanical performance, in addition to hampering the production of particleboards and causing cutting tools to wear out more quickly (Iwakiri 2005).

3.3 Physical properties of the particleboards

The moisture and bulk density of the particleboards for both types of adhesives used were unaffected by the partial replacement of pine particles with bean waste (Figure 6). The moisture contents varied from 6.2% to 6.6%, showing no significant differences among the evaluated compositions. NBR 14810–2 (ABNT 2018) delimits moisture ranging from 5% to 11%. For both properties, all particleboard compositions tested were within the range provided by the standard.

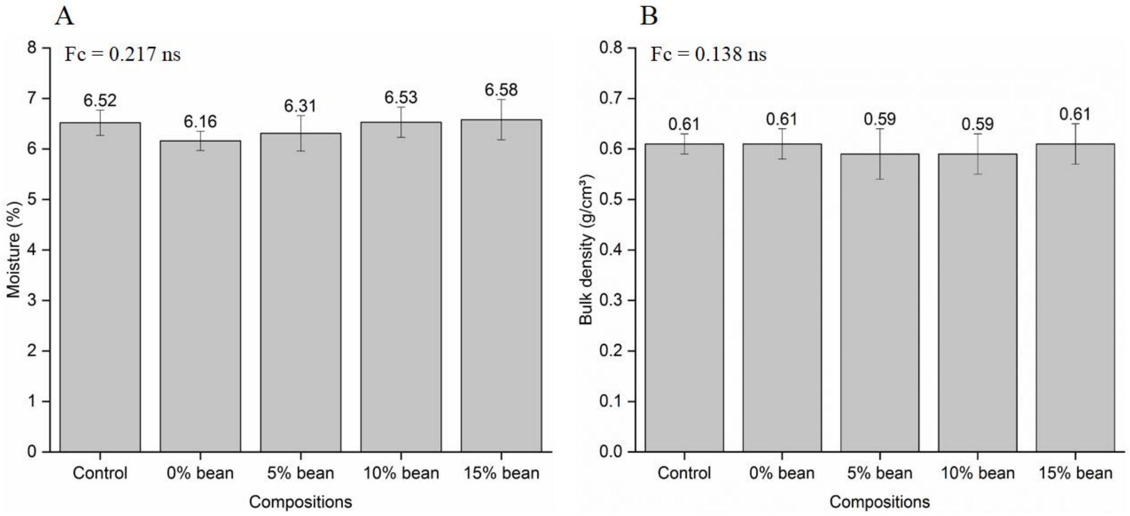

Fig 6 Moisture (a) and bulk density (b) of the particleboards; ns = nonsignificant ($p \geq 0.05$)

The particleboards produced showed bulk densities ranging from 0.59 to 0.61 g/cm³, classified as low density (< 0.64 g/cm³) according to the ANSI A208.1 (2016) standard. This classification is necessary because bulk density is related to the minimum values of thickness swelling, water absorption, modulus of elasticity, modulus of rupture, and internal bond

(Machado et al. 2017). The nominal density (0.65 g/cm³) was higher than the bulk densities due to laboratory circumstances, such as particle losses during sizing, particle mattress formation, and subsequent pressing steps, namely, the spring back that occurs at the press opening, due to stress release and eventually incomplete resin cure (Gonçalves et al. 2020; Faria et al. 2021).

The vertical density profile of particleboards fundamentally affects their resulting mechanical properties (Hýsek and Zółtowska 2022). An effective distribution of the glued-on particles is desirable, promoting the optimal distribution of material along with the profile of the board (Figure 7).

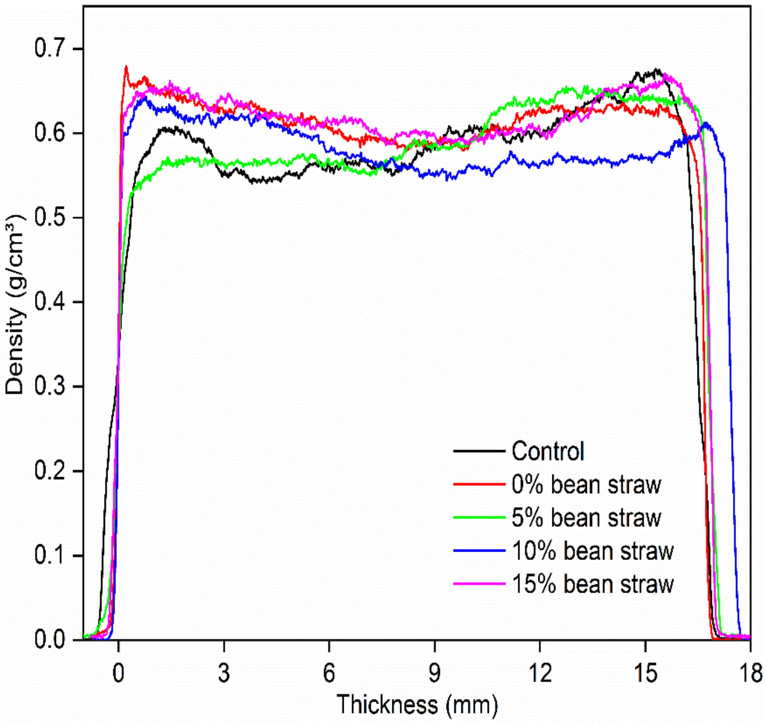

Fig 7 Vertical density profile of the particleboards

The density profile describes the particleboard's density in the layers that compose it along with its cross-section (Gonçalves et al. 2018). In this way, an "M" shaped profile, with higher density values at the faces and lower density values inside the particleboards, is typically observed and depends mostly on the pressing cycle (press closing speed) and moisture content. It can also be because of the type and characteristics of the material used and may affect their physical-mechanical properties (Yemele et al. 2008; Lopez et al. 2021; Korai 2021).

The vertical density profile was very close to the determined nominal density (0.650 g/cm³) of the boards. The densitometry method observed a reduction in density values due to material loss during panel production and spring back after pressing (Kwon and Geimer 1998; Brito and Bortoletto Junior 2020). The particleboards made with a 15% replacement of pine wood particles by bean straw had the highest compression ratio, which was significantly different from the control and 0% bean compositions tested (Figure 8). This behavior is explained by the low basic density of bean waste (0.123 g/cm³), which results in a greater amount of particles needed to reach the established density, resulting in greater compaction and reduction of voids after pressing.

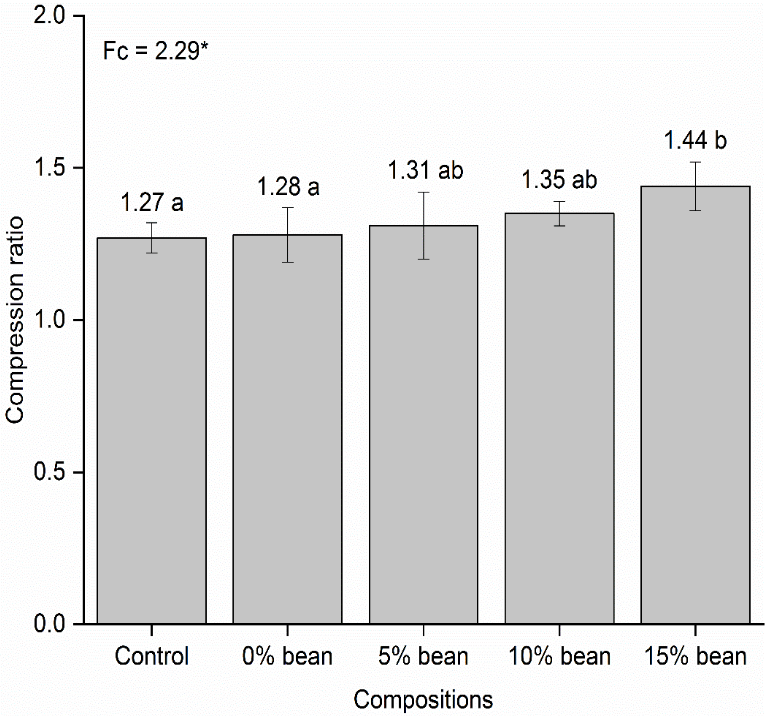

Fig 8 The compression ratio of the particleboards. Averages followed by the same letter indicate no significant difference according to the LSD test ($p > 0.05$). *Significant ($p \leq 0.05$)

The compression ratios of the particleboards made with pine wood and the two types of adhesives (urea and cardanol-formaldehyde) were statistically identical. By increasing the content of bean straw particles in place of pine wood, the particleboard compression ratio increased. The ideal compression ratio ranges from 1.3 to 1.6, according to Maloney (1993). Therefore, only particleboards made with substitution contents of 5%, 10%, and 15% were in the established range. Several authors observed an increased compression ratio as a higher percentage of nonwood lignocellulosic wastes were inserted into the particleboard. According to Guimarães et al. (2019), the compression ratio increased for low-density particleboards made with 0% and 50% soy wastes instead of eucalyptus wood, from 1.5 to 2.2, respectively.

Scatolino et al. (2019) showed that the compression ratio for particleboards increased from 1.2 to 1.4 when cotton waste was used to replace 30% of eucalyptus wood particles.

The substitution of 5% pine wood by bean waste particles resulted in increased thickness swelling at 24 h, not differing statistically from the other compositions (Figure 9).

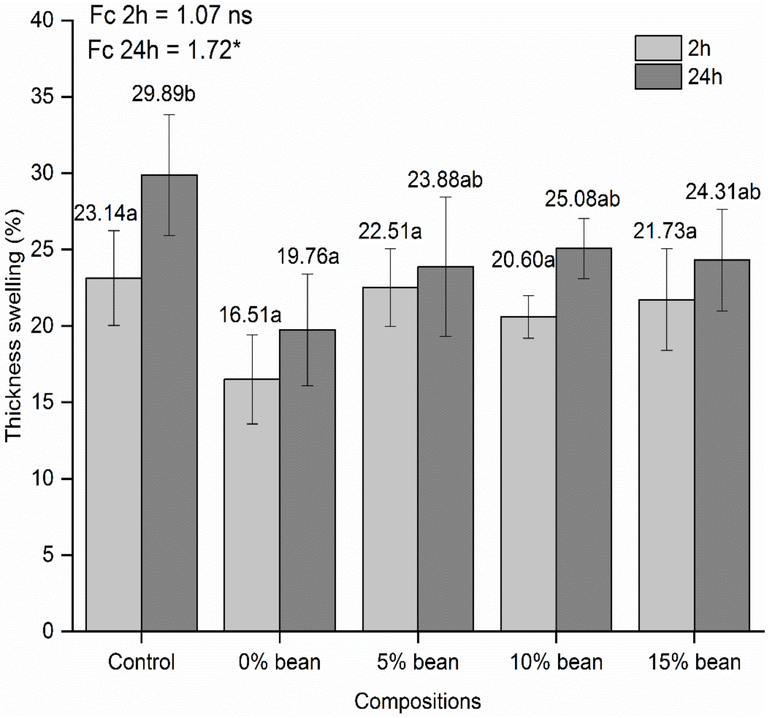

Fig 9 Thickness swelling after 2 and 24 h of immersion. Averages followed by the same letter indicate no significant difference according to the LSD test ($p > 0.05$). *Significant ($p \leq 0.05$); ns = nonsignificant ($p \geq 0.05$)

There were no significant differences in thickness swelling after 2 h of immersion in water among the compositions studied. In terms of thickness swelling at 24 h, particleboard made with pine wood particles (0% bean wastes) had lower average values than those made with urea-formaldehyde adhesive (Control). Despite the increase in compression ratio, the increased number of bean particles required to achieve the nominal density did not affect the particleboard's dimensional stability. As a result, the adhesive coating of the particles was satisfactory, resulting in adhesive bond efficiency (Veloso et al. 2020b).

The chemical composition of the bean wastes also explains the behavior observed in the present work. A high content of total extractives (22%) was verified. Extractives from lignocellulosic material are responsible for reducing its permeability and hygroscopicity because they are hydrophobic compounds with low molecular weights (Iwakiri 2005). The higher lignin content obtained from bean waste may also have influenced the thickness swelling results because of the amorphous, highly branched structure and predominantly aromatic compound (Klock 2013). The lower content of holocellulose (fraction cellulose + hemicelluloses) observed for bean waste (47%) compared to pine wood (70%) was also responsible for the better dimensional stability of the material. Because natural fibers contain cellulose and hemicelluloses, higher concentrations of these compounds influence water absorption and swelling (Scatolino et al. 2019).

Furtini et al. (2022) obtained the opposite result for dimensional stability for particleboards with cardanol-formaldehyde. The authors noticed an increase in thickness swelling after 2 and 24 h of immersion, as higher contents of cardanol in substitution of urea-formaldehyde. A variation was observed in thickness swelling after 2 h immersion in water from 19% to 43% for panels made with substitution of 0% and 80%, respectively. For thickness swelling after 24 h immersion in water, contents varying from 23% to 54% were observed for panels produced with substitutions of 0% and 80%, respectively. The improvement in dimensional stability compared to that found by Furtini et al. (2022) is because they substituted urea-formaldehyde for cardanol by carrying out its synthesis. In this way, covalent bonds were formed between cardanol and formaldehyde, resulting in an efficient bond between the adhesive and particle, with the consequent higher dimensional stability. Wood particles and lignocellulosic wastes can change when exposed to moisture. In some cases, the magnitude of this phenomenon is a limiting factor in using them as raw materials (Machado et al. 2017). The commercial standard ANSI A208.1 (2016) with maximum thickness swelling of 30% after 2 h immersion for low-density boards produced with urea-formaldehyde. In conclusion, given the results and based on the points discussed despite the trends, all the particleboards met the regulatory requirements.

3.4 Mechanical properties of the particleboards

Internal bond are often considered one of the more significant mechanical properties of particleboards (Cesprini et al. 2022). Using a cardanol-formaldehyde adhesive instead of urea-formaldehyde resulted in an increased internal bond strength of 53% for the particleboards made with pine (Figure 10).

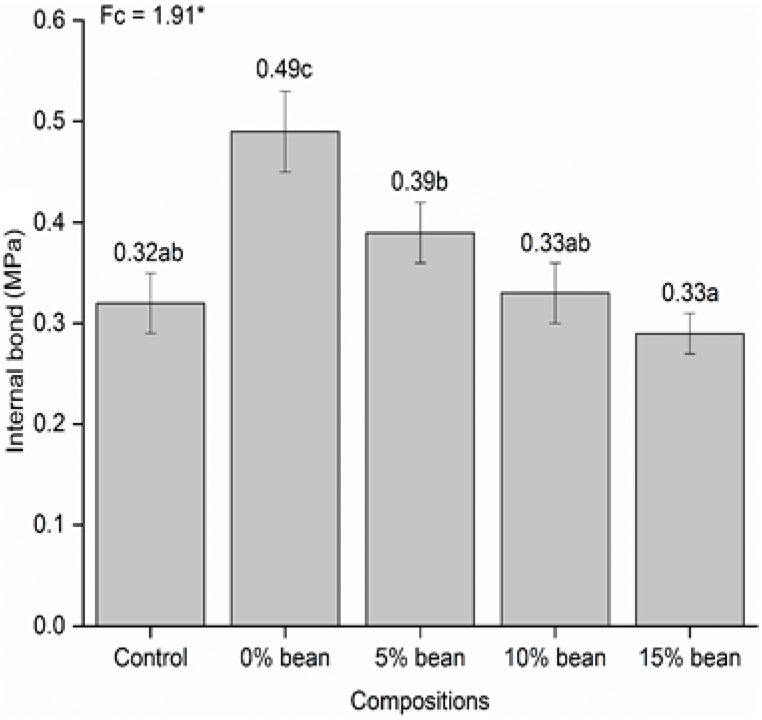

Fig. 10 Internal bond of the particleboards. Averages followed by the same letter indicate no significant difference according to the LSD test ($p > 0.05$). *Significant ($p \leq 0.05$)

Furtini et al. (2022) cited that a 20% replacement of urea with cardanol reduced the internal bond by 25%. However, for particleboards made with phenol-formaldehyde and liquid aldehyde from cashew nuts, Tomkinson et al. (2003) noted that the internal bond increased from 0.69 MPa to 1.05 MPa. As it is a patent, information such as pressing temperature and other process variables that help assess the particleboard's technical and financial viability were not reported. The improvement in the internal bond of particleboards with cardanol-formaldehyde was caused by the cross-linking of formaldehyde with the aromatic groups of cardanol, constructing a hardened network (Pizzi 2006).

Higher contents of bean straw wastes used as partial substitution of pine particles (Figure 10) show a decrease in the values of internal bonding in produced particleboards. The chemical composition may have influenced the interaction between particle and adhesive, mainly due to the high content of extractives, causing inhibition of the particle-adhesive interaction process (Boa et al. 2014). Scatolino et al. (2019) reported an increased compression ratio and a reduced internal bond with higher contents of cotton waste used for partial substitution of eucalyptus in particleboards. The authors observed extractive contents of 22% for cotton wastes and 5.1% for eucalyptus wood. The high concentration of extractives in cotton waste promoted a reduction in internal bonds, which ranged from 0.26

MPa (panels produced without cotton waste) to 0.16 MPa (30% of cotton waste). Micrographs obtained by SEM show the absence of adhesive and an increase in porosity in regions of the particleboards produced with 15% bean straw residues compared to the Control (Figure 11).

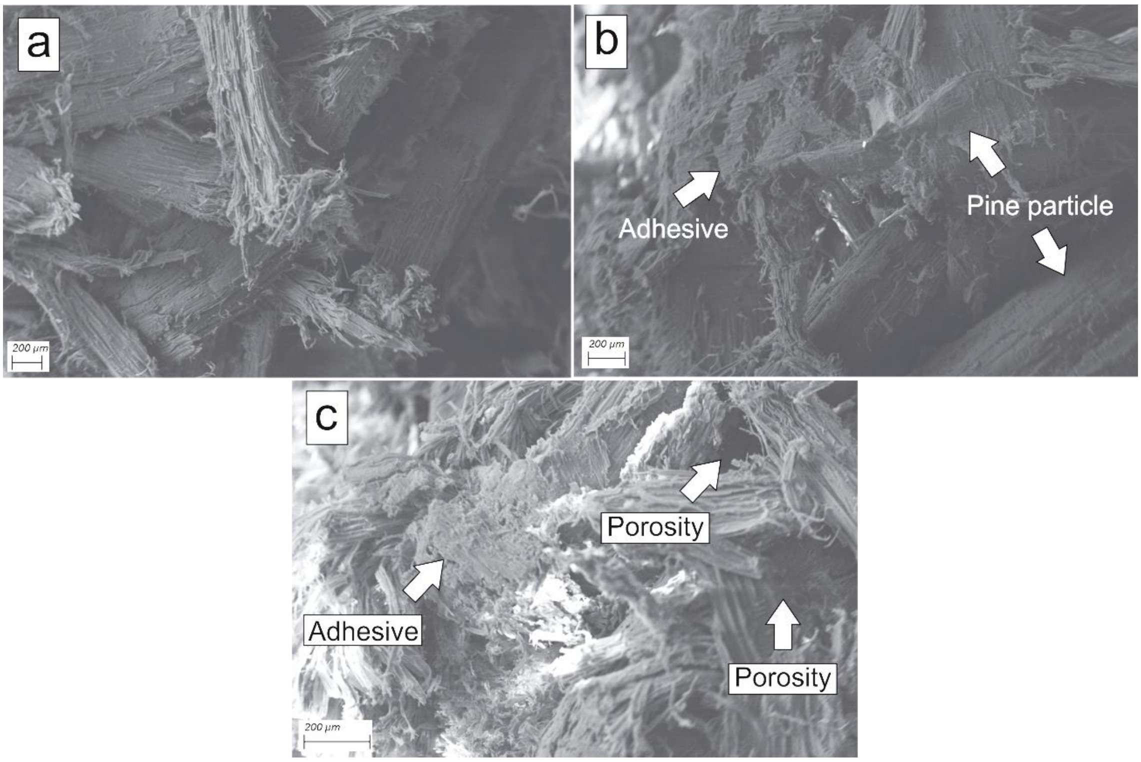

Fig 11 Micrographs of the particleboards; (a) Control; (b) 0% bean straw; and (c) 15% bean straw

The micrographs show that the distribution of adhesive on the particle surface was not uniform (Figure 11c). This fact may have been influenced by the extractives present in the bean residues. Due to the poor distribution of adhesive, a microstructure containing high porosity is observed, directly resulting in the reduction of the strength of the panels. Although more bean residues caused a reduction in the interaction of the cardanol-formaldehyde adhesive with the particles, all the compositions tested met the ANSI A208.1 (2016) standard minimum internal bond value of 0.10 MPa for particleboard made with urea-formaldehyde. Thus, the results indicate that cardanol-formaldehyde has the potential to replace urea-formaldehyde in boards, promoting the interaction of adhesive-particle up to 15% of bean wastes inserted.

When compared to other compositions, the particleboards made with cardanol-formaldehyde showed a significantly different MOR. The substitution of urea-formaldehyde for cardanol-formaldehyde caused an increase from 2.45 MPa to 4.39 MPa for MOR (Figure 12).

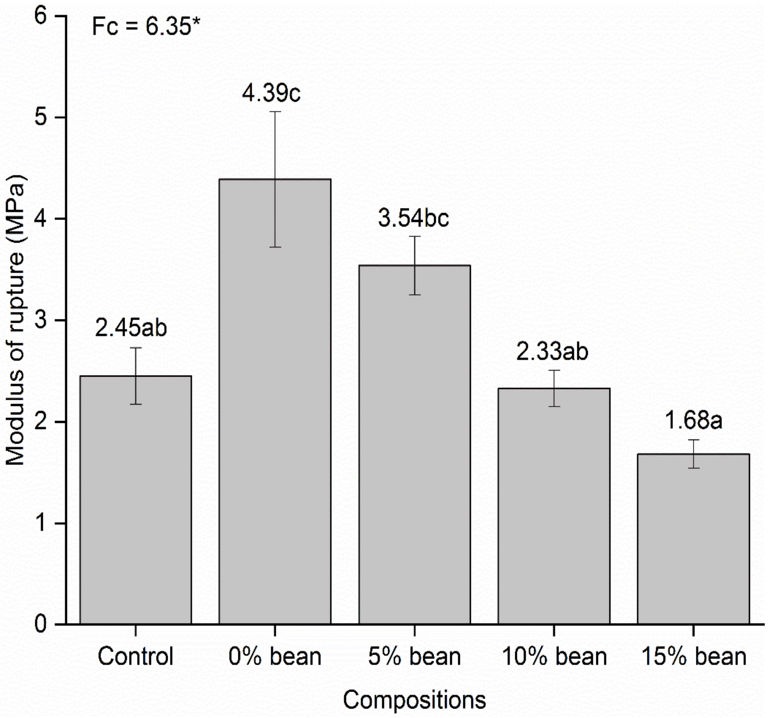

Fig. 12 Modulus of rupture in the static bending test. Averages followed by the same letter indicate no significant difference according to the LSD test ($p > 0.05$). *Significant ($p \leq 0.05$)

Figure 12 shows the MOR reduction behavior by increasing the bean straw particle content when comparing the panels produced with cardanol-formaldehyde. This reduction is primarily due to the high amount of extractives present in the wastes, which causes a bonding deficiency (Fonte and Trianoski 2015). Furthermore, the extractives may have migrated to the surface and concentrated in large quantities during the drying of the particles, inactivating the surface and weakening the contact with the adhesive (Iwakiri 2005).

The modulus of elasticity (Figure 13) followed the same trend observed for the modulus of rupture. Compared to the control particleboard, by using cardanol-formaldehyde, 0% and 5% bean resulted in a significant increase in MOE. Compared with the control particleboard, the use of 5% bean straw resulted in a significantly higher MOE value. However, a statistically lower MOE value than the control composition was observed for panels produced with 15% bean straw. The reduction in the values observed for MOR and MOE may be due to the higher compression ratio since there is a greater number of particles and, consequently, an increase in the surface area. In this way, using the same amount of adhesive will reduce its availability per particle, which results in a decrease in mechanical properties (Faria et al. 2022). The use of cardanol-formaldehyde promoted an increased MOR by 79% and the MOE by 53% for the particleboards made with pine wood.

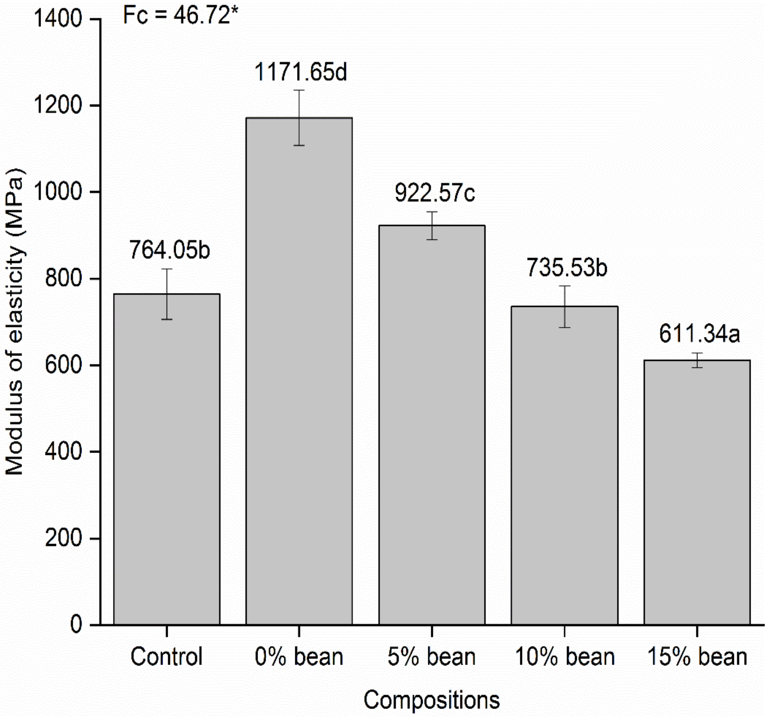

Fig. 13 Modulus of elasticity in the static bending test. Averages followed by the same letter indicate no significant difference according to the LSD test ($p > 0.05$). *Significant ($p \leq 0.05$)

The ANSI A208.1 (2016) commercial standard establishes minimum values of 550 MPa for MOE and 3.0 MPa for MOR for low-density particleboards glued with urea-formaldehyde. All compositions presented values higher than these levels for MOE. For MOR, only the boards produced with 0% and 5% pine wood replacement by bean waste comply with the regulation.

3.5 Combustion properties of the particleboards

The combustion experiment of particleboard specimens conducted as a function of burning time is shown in Figure 14. For each stage of burning, the flame increased until a large part of the particleboard was consumed.

Fig. 14 Fire stages during the combustion of the particleboards; a) 1 min; b) 3 min; c) 5 min; d) 10 min

In comparison to the other compositions, the particleboards made with urea-formaldehyde (Control) showed a larger mass loss in the initial minutes. However, it was verified that the results from the different compositions were very close (Figure 15). Similar behavior was observed by Furtini et al. (2022), in which the authors found more significant mass loss for particleboards with urea-formaldehyde. The chemical composition of cardanol influenced the resistance to combustion, mainly because of the presence of the aromatic ring and the position of the double bonds, thus requiring more energy for thermal degradation to occur.

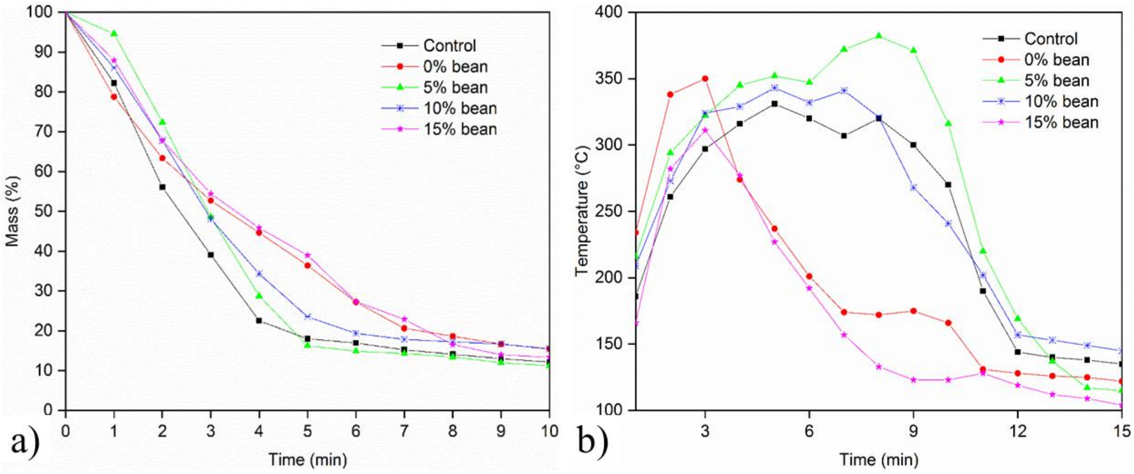

Fig 15 a) Mass variation with combustion time; b) Temperature variation with combustion time

Even though the cardanol-formaldehyde adhesive is more resistant to combustion, the addition of bean waste reduced the particleboard's thermal stability. This reduction can be attributed to the chemical composition of bean wastes, which contained more extractives and less lignin. Depending on the type and content of the extractives, they affect thermal degradation, promoting lignocellulosic material flammability at lower temperatures. Accelerating the degradation of one component can expedite the degradation of the material's other components (Poletto et al. 2012; Valette et al. 2017).

Particleboards are composites produced with particles joined by adhesive under pressure and temperature (Iwakiri 2005). Therefore, the particles' chemical characteristics strongly influence the board's combustibility. At temperatures below 200 °C, the degradation of low molar mass compounds, such as lipids, fatty acids, resin acids, and waxes, present in extractives occurs (Floch et al. 2015). Hemicellulose degradation occurs between 200 and 315 °C, followed by cellulose degradation between 300 and 400 °C. Finally, lignin degradation occurs from 300 to 600 °C (Floch et al. 2015; Le van 1992; Bianchi et al. 2010).

In the first minute of testing, the temperature exceeded 250 °C for all compositions evaluated (see Figure 15b). The degradation of extractives and hemicelluloses has already begun in this temperature range. In other words, the higher the extractive content is, the faster the initial degradation of the particleboards, thus increasing the combustion temperature because of the high calorific value of these components (Guo et al. 2010; Telmo and Lousada 2011). Therefore, the higher the combustion temperature in the first minutes, the faster the degradation of the other chemical components of the cell wall. This explains the abrupt decrease in temperature for the particleboards with 15% bean waste. In addition, the lower lignin content may have influenced the degradation of the panels since this component has a higher calorific value due to the carbon-carbon bonds between the monomeric phenyl-propane units, in addition to the higher carbon content of this molecular component of the biomass (Howard 1973; Demirbas 2001; Sharma et al. 2004).

3.6 Formaldehyde emission

Particleboards produced with cardanol-formaldehyde showed an intense reduction in formaldehyde emission values compared to those produced with urea-formaldehyde (Table 3). A 93% reduction in formaldehyde emission values was observed for particleboards made with cardanol-formaldehyde (0% bean) compared to those produced with urea-formaldehyde (Control). Furthermore, the water containing formaldehyde presented a yellowish color for the particleboards with urea-formaldehyde (Control) (Figure 16). In contrast, a transparent color was verified for the particleboards with cardanol.

Table 3 Emission of formaldehyde from particleboards studied according to EN 717-3 (1996) (flask method)

Composition	Adhesive	Formaldehyde release (mg/100 g oven dry-board)
Control	Urea-formaldehyde	16.76
0% bean	Cardanol-formaldehyde	1.09
5% bean	Cardanol-formaldehyde	1.41
10% bean	Cardanol-formaldehyde	1.15
15% bean	Cardanol-formaldehyde	1.30

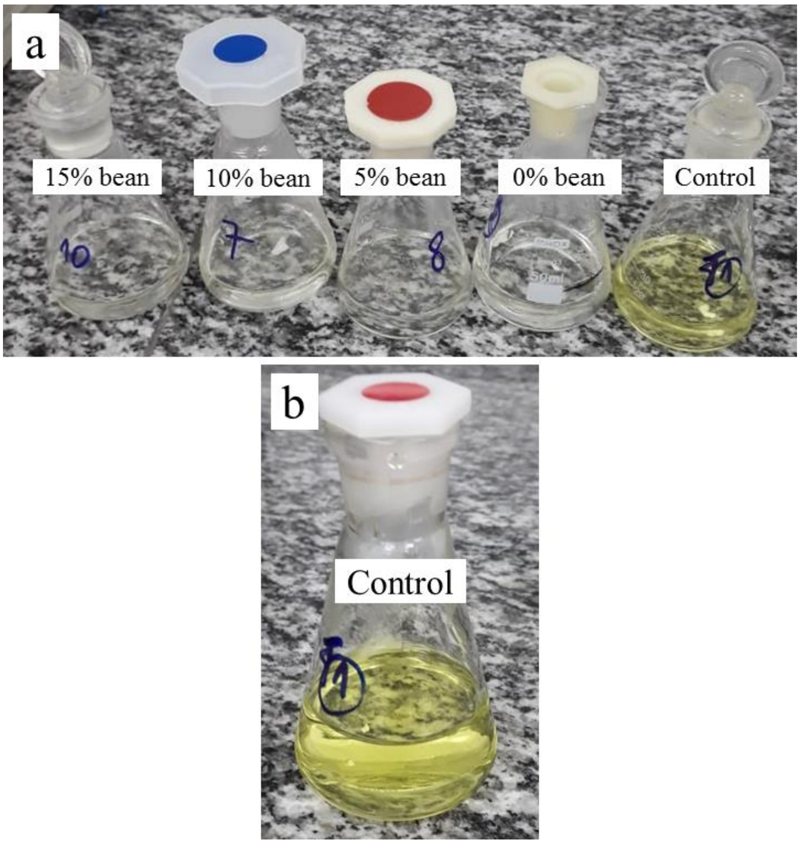

Fig. 16 a) Formaldehyde absorption in water for the different particleboards produced; b) details of formaldehyde-containing water for the particleboards made with urea-formaldehyde

In terms of formaldehyde emission, all particleboards made with cardanol-formaldehyde were categorized in class E1 (< 8 mg/100 g). In comparison, boards made with urea-formaldehyde were categorized in class E2 (8–30 mg/100 g) (EN 13986+A1 2015). The methylene bridges (CH_2) that totally or partially surround the phenolic fractions and are more stable than the methylene bridges of urea-formaldehyde are responsible for the reduction in formaldehyde emission of the cardanol-formaldehyde samples (Younesi-Kordkheil et al. 2015). Bisanda et al. (2003) showed a decrease in formaldehyde emission values in particleboards and confirmed that the environmentally friendly adhesive tannin-cardanol-formaldehyde emits less formaldehyde than urea-formaldehyde. The potential of cardanol-formaldehyde as an adhesive could be attractive in board industries. In addition to complying with marketing standards regarding physical and strength properties, it also provides lower formaldehyde emission properties, which is an advantage in consumer health issues.

Conclusions

Because of the intrinsic characteristics of cardanol, such as a long and saturated chain (C15) and the presence of an aromatic ring, the technological properties of the particleboards were influenced. Particleboards glued with cardanol-formaldehyde and pine wood showed the best mechanical performance and reduced water absorption compared to panels made with urea-formaldehyde. The formaldehyde emission (mg/100 g oven-dry board) was reduced by 93% in the boards produced with urea-formaldehyde and cardanol-formaldehyde. The insertion of up to 15% of bean wastes increased water absorption levels after 2 and 24 h of immersion, in addition to promoting a reduction in the MOR, MOE, and formaldehyde emission by 93%. Cardanol-formaldehyde proved to be a potential adhesive used in wood panel industries, meeting the requirements of commercialization and use indoors. The final product still needs to be properly prepared for outdoor environments. The properties of these panels could be improved by adding a treatment that covers the particles with some material that has hydrophobic characteristics or resorts to some residue that has, in its composition, extractives with water repellency.

Acknowledgments

We are especially grateful to the Postgraduate Program in Biomaterials Engineering (PPGBIOMAT) of the Federal University of Lavras (UFLA) for providing material and infrastructure. The authors are also grateful to the National Council for Scientific and

Technological Development (CNPq finance code 300985/2022). We also thank the Fundação de Amparo à Pesquisa do Estado do Amapá (FAPEAP) for the postdoctoral scholarships (financial code: 0022.0279.1202.0016/2021 – PROPESP).

Funding information

The authors received no financial support for the research, authorship, and/or publication of this article.

Availability of data and material

The datasets supporting the conclusions are included in the manuscript. Furthermore, the datasets analyzed in this study are available from the corresponding author upon request.

Code availability

Not applicable

Authors' contributions

Douglas Lamounier Faria: Conceptualization, Investigation, Data Curation and Writing – original draft.

Mário Vanoli Scatolino; Juliano Elvis de Oliveira; Julio Soriano; Thiago de Paula Protásio and Luisa Maria Hora de Carvalho: Conceptualization; Methodology; Validation; Writing - original draft.

Fabricio Gomes Gonçalves and Roberto Carlos Costa Lelis: Investigation; Methodology; Validation; Writing - original draft.

Lourival Marin Mendes and José Benedito Guimarães Junior: Funding acquisition, Supervision, Resources, Project administration.

Conflicts of interest

The authors declare they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

Ethical Approval

Not applicable. This manuscript does not involve researching humans or animals.

Consent to Participate

All of the authors consented to participate in the drafting of this manuscript.

Consent to Publish

All of the authors consented to publish this manuscript.

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