



Cite this: *RSC Sustainability*, 2025, **3**, 376

Received 21st August 2024
Accepted 6th November 2024

DOI: 10.1039/d4su00503a
rsc.li/rscsus

Mechanically processed Sardinian wool promotes C–C bond synthesis under solvent-free conditions[†]

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The tangible environmental concerns of the last few decades are prompting science to a new sustainable paradigm for chemical reactions and a circular economy. In the present manuscript, complying with this dichotomy, Sardinian wool is presented as a promising material for organic synthesis. Aldol and nitro-aldol additions, and Knoevenagel reactions were demonstrated with high efficiency under wool-promoted and solvent-free conditions, paving the way to novel approaches for converting wool biomass waste into value-added products for C–C bond formation.

Sustainability spotlight

Our research on Sardinian wool aligns with the goal of a carbon-neutral society by repurposing an undervalued natural resource into sustainable materials. Wool, as a biodegradable and renewable resource, offers an eco-friendly alternative to synthetic fibers and energy-intensive materials. By focusing on the development of low-impact applications, such as barely solvent-free pre-treatments and solvent-less reactions, we aim to reduce the reliance of organic chemistry on toxic, expensive and/or volatile catalysts and chemicals. This approach not only supports waste reduction but also leverages local resources, contributing to regional economic sustainability. Our work addresses, directly and indirectly, some UN sustainable development goals: industry, innovation, and infrastructure (SDG 9), responsible consumption and production (SDG 12), and climate action (SDG 13).

Introduction

The increasing need for environmental preservation has highlighted the importance of sustainable practices in chemistry, leading to the development of green chemistry.¹ This field, governed by 12 principles,² emphasizes the use of renewable feedstocks to reduce reliance on non-renewable resources.³ Green chemistry aims to design chemical products and processes that minimize the use and generation of hazardous substances.⁴ The valorisation of plant- and animal-based waste, then, has gained significant attention as a means to enhance recyclability and achieve long-term sustainability.⁵

According to the Global Waste Management Outlook (GWMO), the total municipal solid waste generated globally in 2023 was approximately 2.1 billion tons, originating primarily from household activities, agricultural operations, and industrial and construction activities.⁶ Converting this waste into value-added products is crucial for addressing environmental concerns and promoting sustainability.⁷ Effective waste reduction strategies, improved recycling technologies, and robust policies are essential to promote sustainable consumption and

production practices. Within this context, reusing waste within a circular economy framework, such as that of the European Union, is key to extending the life cycle of materials and products through practices like sharing, leasing, reusing, repairing, refurbishing, and recycling.⁸

Wool, a natural polymer derived from sheep, is valued for its unique mechanical and physical properties.⁹ It primarily consists of keratin, a protein rich in amino acids like cystine, which imparts resilience and desirable physical and mechanical properties.¹⁰ The disposal of wool waste, regulated by EU directives, poses significant challenges,¹¹ especially in regions like Sardinia, which has a high sheep population and generates substantial wool waste.¹²

Recent research has explored using natural fibres as a support for catalysis in various reactions.¹³ With regards to wool, its amino acid composition and isoelectric point make it suitable for binding metal cations, facilitating its use in metal-based catalytic processes.¹⁴ For instance, metal catalysts based on Pd, Fe, Cu, and Mn supported on wool have been successfully utilized in C–C/C–N cross-coupling, desulfuration coupling, and oxidation reactions.¹⁵ However, these methodologies often relied on chemical treatment of the fibres and on toxic solvents, such as DMSO and TBAB, which limit their alignment with green chemistry principles.¹⁶ Addressing these limitations is crucial for developing truly sustainable catalytic processes.

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[†] Electronic supplementary information (ESI) available. See DOI: <https://doi.org/10.1039/d4su00503a>



Inspired by the catalytic activity of amino acids and proteins in aldol and nitroaldol reactions,¹⁷ we envisaged amino acid-containing fibers as potent promoters of organic transformations.

In this study, we report using Sardinian wool powder as a sustainable and efficient promoter for aldol, nitroaldol, and Knoevenagel reactions. By employing a ball milling device, a consolidated enabling technology for green chemistry applications,¹⁸ to enhance surface area and catalytic activity, our approach aligns with the principles of green chemistry by operating under solvent-free conditions during the reaction. This approach valorizes wool waste and contributes to the circular economy of Sardinia, transforming waste into a valuable material for catalysis.

Results and discussion

FT-IR analysis

All the wool samples subjected to different treatments were analysed using FT-IR spectroscopy. Spectrum A (Fig. 1) corresponds to the black wool samples, whereas Spectrum B (Fig. 1) corresponds to the white wool samples. In Spectrum A, the FT-IR data for black wool fiber (BWF), black wool powder (BWP), and

black wool powder treated with 10% HCl solution (BWP(HCl)) have been compared. In Spectrum A, we observed a strong band at 1657 cm^{-1} for C=O stretching frequency in all the black wool samples, indicating the presence of α -helix structures. Furthermore, at 1543 cm^{-1} , peaks were observed, particularly Amide-II bands of noncyclic secondary amides arising from the combined effect of N-H bending and C=N stretching frequencies. A strong band was also observed at 1635 cm^{-1} corresponding to Amide-I C=O stretching frequency for β -pleated sheets. These observations indicate the presence of both α -helix and β -pleated sheets in the black wool samples. Among all the black wool samples, these peaks remain unchanged, indicating the retention of the secondary structure in the wool samples after mechanical treatment (milling in ball mills) and chemical treatment (treatment with acid). In Spectrum B, we observed similar outcomes in FT-IR analysis. Herein, the Amide-I C=O stretching frequency for α -helix structures was observed at 1540 cm^{-1} , and the Amide-I C=O stretching frequency for β -pleated sheets was observed at 1635 cm^{-1} . These wool samples were also resistant to changes in secondary structures upon mechanochemical and chemical treatments.

Aldol reactions

We commenced our investigation on the evaluation of wool performances by testing a model reaction. We selected the aldol addition between cyclopentanone **1a** and 4-nitro benzaldehyde **2a** to obtain the aldol product **3aa** in the presence of white wool powder. The best conditions were found to be as follows: **2a** (0.20 mmol), **1a** (1.00 mmol), water (98 μL), and 50 mg of white wool powder gently stirred at r.t. for seven days (98% yield, Table 1, entry 1). Deviations from the standard conditions, involving chicken feather powder (FP), are reported in Table 1 and discussed below.

Initially, we fine-tuned the best stoichiometric ratio for the aldol reactions, which we found to be good in 10-fold excess of the ketone (entry 2, Table 1). However, since no significant decrease in yield was observed by halving this amount, we opted for a 1 : 5 ratio for waste prevention purposes (entry 1, Table 1). Further reduction in the amount of ketone resulted in poor results compared to the previously mentioned ones. Notably, decreasing the amount of white wool powder from 50 mg to 25 mg resulted in a substantial decrease in conversion into **3a** from 98% to a mere 47% (entry 3, Table 1), suggesting that 50 mg of WWP is the minimum amount that is needed for this kind of chemical transformation. Comparable results were obtained when white wool powder was replaced by black wool powder or chicken feather powder, further supporting the role of keratin powder in the reaction (entries 4 and 5, Table 1). However, with the use of WWP(HCl) instead of WWP, the yield of product **3a** saw a remarkable drop (entry 6, Table 1), suggesting that side chains containing ionizable N-residues are crucial for the reaction to occur, further validating the mechanism *via* enamine catalysis. With the reduction of reaction time from 7 days to 4 days, the conversion of product **3a** decreased to 61% (entry 7, Table 1). Furthermore, the reaction in the presence of WWF instead of WWP had inferior yields compared to

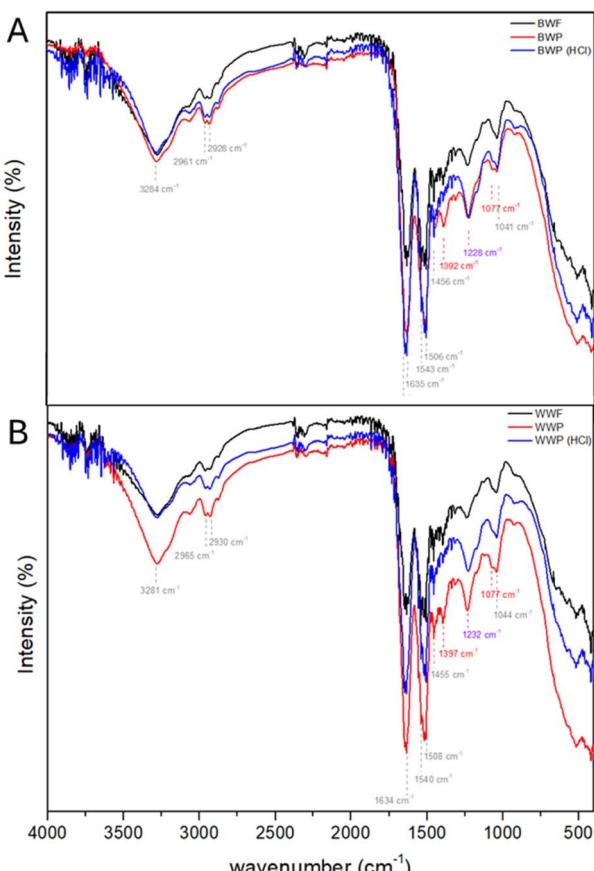
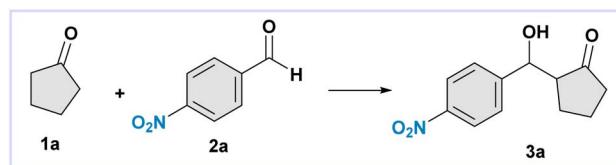


Fig. 1 FT-IR spectra of wool samples. Spectrum A: black wool fiber (BWF), black wool powder (BWP), and acid-treated black wool powder (BWP(HCl)). Spectrum B: white wool fiber (WWF), white wool powder (WWP), and acid-treated white wool powder (WWP(HCl)).

Table 1 Optimization and screening of the keratin-catalyzed aldol reaction

	Deviation from standard conditions	3a (NMR yield)
1	None ^a	98% ^b
2	10 mmol of 1a instead of 5 mmol	99%
3	25 mg of wool instead of 50 mg	47%
4	BWP instead of WWP	93% ^c
5	FP instead of WWP	95% ^d
6	WWP(HCl) instead of WWP	23%
7	4 days instead of 7 days	61%
8	WWF instead of WWP	7%
9	1 mL of H ₂ O instead of 98 μ L	58%
10	DMSO instead of H ₂ O	—
11	EtOH instead of H ₂ O	—
12	AcOEt instead of H ₂ O	12%
13	40 °C instead of r.t.	80%
14	No WWP ^e	—

^a Reaction conditions: **2a** (0.20 mmol), **1a** (1.00 mmol), water (98 μ L), and 50 mg of white wool powder were gently stirred at r.t. for 7 days.

^b Approximate diastereoisomer ratio *anti/syn* = 0.4 : 1. ^c Approximate diastereoisomer ratio *anti/syn* = 0.5 : 1. ^d Approximate diastereoisomer ratio *anti/syn* = 0.5 : 1. NMR yield was calculated using trimethoxybenzene as the internal standard. ^e The reaction has been monitored for 10 days.

1b and **1c** reacted with 4-nitrobenzaldehyde **2a** to obtain the corresponding products **3ba** and **3ca** in excellent yields (96% and 83%, respectively, Scheme 1A). Comparing the reactions performed on black wool, white wool, and feather powders, we achieved almost similar yields and diastereomeric ratio outcomes. Notably, in the case of benzaldehydes **2a** and **2i**, an inverted diastereomeric ratio *syn/anti* = 1 : 0.6 was obtained when using black wool powder instead of white wool powder. Lastly, the present reaction was optimized for gram scale synthesis, and the recyclability of wool was assessed, showing that the material can be reused for up to 5 complete cycles despite a progressive loss in yield and material, likely due to material poisoning.

Nitro-aldol reactions

Based on our investigation regarding the aldol reaction, we investigated the nitro-aldol response using the same 4-nitro benzaldehyde **2a** and nitromethane **4** as the model substrates. Through the initial screening, we found that the reaction gave the best results with a 10-fold excess of nitromethane compared to benzaldehyde to achieve the nitro-aldol product after 24 h. A library of compounds (**5a–5m**, 30–95%) was synthesized under the optimized conditions (Scheme 1B). The reactions provided excellent yields in the presence of electron-withdrawing groups on the benzaldehyde ring (**2a–f**). With the substitution at the *meta* position, we obtained only a slight decrease in yields compared to the *para*-substituted benzaldehydes. Black wool and chicken feathers were also used to assess the products, which showed comparable outcomes. The kinetic trend of this reaction was obtained by GC-MS, providing the kinetic curve in Fig. 2 below.

WWP (entry 8, Table 1), suggesting the significance of the mechanical treatment of wool fibers. This can be explained by the increased active sites and surface area generated upon grinding the keratin fibres. At the same time, the primary and secondary structures of the protein are preserved, as shown in the IR spectra of the treated samples (Fig. 1). Finally, different solvent conditions and a controlled temperature of 40 °C did not provide any significant advantage (entries 9–13, Table 1), likely due to some a hydrophilic–hydrophobic balance between wool powder and reagents.

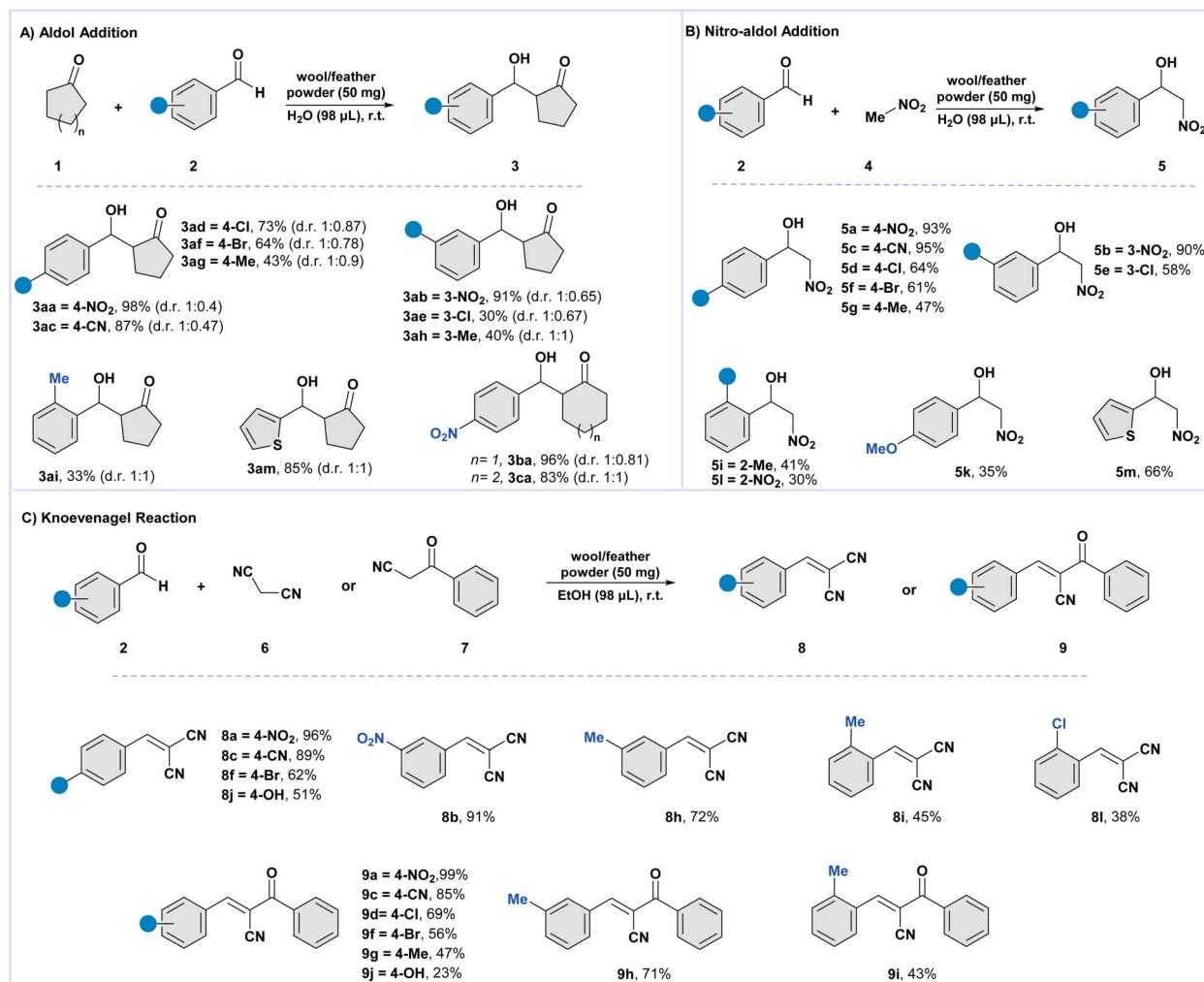
Based on the standard optimized conditions, we decided to test the reaction against differently substituted benzaldehydes. Compounds with electron-withdrawing groups and heterocyclic aldehyde, as in the case of **2a–c** and **2m**, reacted smoothly to provide excellent yields (85–98%, Scheme 1A). Halogen-bearing benzaldehydes like **2d**, **2e**, and **2f** provided the corresponding products in yields ranging from acceptable to good (30–73%, Scheme 1A). Additionally, the effect of the position of the electron-withdrawing group on the benzaldehyde ring from *para* to *meta* positions yielded comparable yields. However, in the case of aldehyde **2e**, we observed a significant drop in yield compared to its *para*-substituted counterpart with **2d**. Notably, benzaldehydes substituted with electron-donating residues, namely **2g** and **2h**, provided the aldol product in acceptable yields (43% and 40%, respectively, Scheme 1A). Lastly, ketones

Knoevenagel reaction

Under our previously optimized conditions, we decided to focus on the Knoevenagel reaction using 4-nitro benzaldehyde **2a** and dicyanomalonate **6** in the presence of WWP. Notably, the response was investigated using a stoichiometric ratio of the two reagents to avoid any purification process involving column chromatography or crystallization of the reaction mixture. This entailed extending the reaction time up to 14 days for completion, however remarkably improving the waste and toxicity prevention. Since the aldol adduct intermediate must undergo a dehydration process to form the π bond, a different solvent in place of H₂O has been investigated. The solvent screening is reported below in Table 2. We found a good compromise between efficiency and greenness with EtOH (entry 5, Table 1), so all the reactions were carried out in the presence of 98 μ L of EtOH. Generally, polar solvents work better than apolar ones; specifically, polar protic solvents lead to higher product yields of **8a**.

Based upon the optimized conditions, the reaction scope was extended to differently substituted benzaldehydes. Compounds bearing electron-withdrawing groups, as in the case of **2a–c**, provided excellent yields of products **8a–c** (87–98%, Scheme 1C). Benzaldehydes having halogens, like **2f** and **2l**, provided the corresponding products **8f** and **8l** in excellent





Scheme 1 Reaction scope for wool-catalyzed organic reaction models. Section 1A: the aldol reaction scope. Section 1B: the nitro-alcohol reaction scope. Section 1C: the Knoevenagel reaction scope.

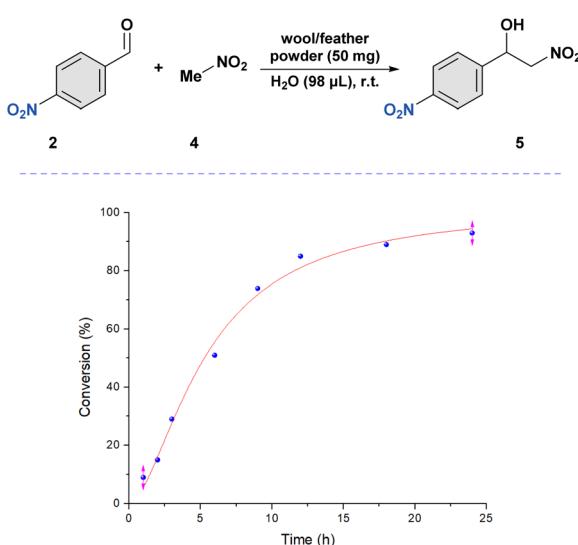


Fig. 2 Reaction rate followed for the nitro-alcohol reaction under wool-catalyzed conditions. GC-MS was used to calculate the conversion (%).

and acceptable yields (62% and 38%, respectively, Scheme 1C). Instead, benzaldehydes bearing electron-donating residues, namely **2h-j**, provided the aldol product in good yields (45–51%, Scheme 1C). A similar reaction trend was observed when benzoyl acetonitrile **7** was used instead of malononitrile. A broad library of compounds was obtained in yields ranging from excellent to poor (**9a-j**, 23–99%, Scheme 1C). The reactions with *meta*-substituted benzaldehydes (**2b** and **2h**) resulted in yields comparable to those of their *para*-substitution counterpart. In the case of *ortho*-substituted benzaldehyde (**2i** and **2l**), we observed a significant decrease in the formation of the desired products.

Experimental

Materials and methods

All components of the wool samples were collected from the Sardinian province of Italy. White wool fibers were collected from the city of Ittiri in the northern region of Sardinia, while black wool fibers were obtained from Guspini, a town in the Sardinian inland. The wool was procured from the Sechi farm,

Table 2 Optimization and screening of the keratin-catalyzed nitro-aldol reaction

Entry	Solvent	8a (NMR yield) ^a
1	Hexane	23%
2	Toluene	20%
3	Distilled H ₂ O	8%
4	AcOEt	73%
5	MeOH	96%
6	Absolute EtOH	98%
7	2-MeTHF	58%

^a Reaction conditions: 2a (0.20 mmol), 6 (0.2 mmol), solvent (98 μ L) and 50 mg of white wool powder were gently stirred at r.t. for 14 days. NMR yield was calculated by using trimethoxybenzene as an internal standard.

ensuring full compliance with animal welfare standards. Chicken feathers were obtained from the Concimi-Bio company. All the wool fibers and chicken feathers were obtained as raw materials and then treated in our laboratory. All reagents and solvents were purchased from Sigma-Aldrich, Fluorochem, and TCI. Dimethyl sulfoxide-d6 (99.9% D) and chloroform-d (99.9% D) were also purchased from Sigma-Aldrich and TCI. All purchased chemicals were used without further purification.

Wool and feather sample preparation

The wool samples were prepared using the following procedure: the raw material was thoroughly rinsed and washed with tap water and soap to eliminate hydrophilic impurities. Afterward, the fibers were rinsed with heptane and then with acetone to remove the hydrophobic impurities. After several washes, the fibers were dried overnight inside an oven. To prepare the acidic wool samples, 10 g of washed wool fibers were placed inside a beaker containing 500 mL of HCl solution (10%) and left untouched for 72 hours. Afterward, the treated fibers were washed with regular tap water and dried overnight in an oven. The dried long fibers have been finely cut into 1 cm pieces to obtain the desired wool fiber (WF) size. The cut fibers were ground with ball mills using zirconia jars and balls. The best results in terms of finer powder were achieved using a Fritsch P7 Premium planetary ball mill (3.00 g of cut fiber inside a 20 mL zirconia jar with 35 balls with diameter Φ = 5 mm and mtot = 13.75 g). After grinding, black wool powder (BWP) and white wool powder (WWP) have been obtained. Wool powders were also obtained from the hydrochloric acid-treated wool fibers (BWP(HCl) and WWP(HCl)) using the same ball milling methodology.

General procedure for synthesis of nitro-aldol compounds

Inside a 4 mL glass vial equipped with a magnetic stirrer, aromatic aldehyde (0.20 mmol), nitromethane (10 equiv.), and

50 mg of wool powder were placed in 98 μ L of distilled water. When the reaction was completed, mixture was gently stirred, and the reaction progress was monitored using TLC (Heptane/AcOEt 1 : 1). The product was recovered by implementing simple filtration using AcOEt with a paper filter. After that, the solvent was evaporated under reduced pressure to obtain the pure products.

General procedure for the synthesis of aldol compounds

Inside a 4 mL glass vial with a magnetic stirrer, aldehyde (0.20 mmol), ketone (5 equiv.), and 50 mg of wool powder were placed in 98 μ L of distilled water. The mixture was gently stirred, and the reaction progress was monitored using TLC (Heptane/AcOEt 5 : 1) until completion. The product was recovered by implementing simple filtration using AcOEt with a paper filter. After that, the solvent was evaporated under reduced pressure to obtain the pure products.

General procedure for the synthesis of Knoevenagel compounds

Inside a 4 mL glass vial equipped with a magnetic stirrer, aldehyde (0.20 mmol), malononitrile or benzoyl acetonitrile (0.20 mmol), and 50 mg of wool powder were placed in 98 μ L of absolute ethanol. The mixture was gently stirred, and the reaction progress was monitored using TLC (Heptane/AcOEt 5 : 1) until completion. The product was recovered by implementing simple filtration using AcOEt with a paper filter. After that, the solvent was evaporated under reduced pressure to obtain the pure products.

Conclusions

In conclusion, this investigation aimed to test the potential of Sardinian wool as a renewable and sustainable catalyst for various organic reactions. Derived from its keratin content and functional groups, wool's features confer a unique advantage in heterogeneous catalysis. Our findings have shown that wool-based catalysts effectively promote aldol and Knoevenagel reactions, highlighting the versatility and efficiency of wool in catalytic applications. The optimization of the reaction conditions revealed that the stoichiometry, the composition of the catalytic support, and mechanical processing significantly impact the catalytic performances. FT-IR analyses confirmed that the structural integrity of wool is preserved across different treatments, thus maintaining the necessary activity for catalytic purposes. Comparative tests with other keratinous materials, such as chicken feathers, suggested a broader applicability of keratin-based catalysts. Moreover, the use of locally sourced wool not only promotes the valorization of what is traditionally considered agricultural waste but also contributes economically and environmentally to local communities in Sardinia, embodying the ideals of the circular economy.

Data availability

The data supporting this article have been included as part of the ESI.†



Author contributions

F. C. wrote the first draft of the article. F. C. and A. P. conceptualized the idea and revised the manuscript's drafts. F. C. and So. B. performed the syntheses and collected the data. F. C., F. B., and St. B. analysed the data. A. P. supervised the work. All the authors contributed to the draft's correction and finalization.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This study was carried out within the RETURN Extended Partnership and received funding from the European Union Next-GenerationEU (National Recovery and Resilience Plan – NRRP, Mission 4, Component 2, Investment 1.3 – D.D. 1243 2/8/2022, PE0000005). MASE also provided financial support for the project having grant number F33C23000680001 and MUR with PNRR PE2 – NEST – Network 4 Energy Sustainable Transition – (MUR no. PE0000021, CUP no. F53C22000770007). We acknowledge Prof. Angelo Frongia for having shared his expertise in aldol reactions and the CeSAR (Centro Servizi Ricerca d'Ateneo) core facility of the University of Cagliari, Dr Sandrina Lampis for assistance with the generation of NMR data and Dr. Marco Marceddu for assistance with the generation of SEM images.

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