

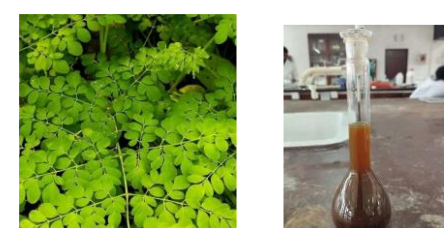
## Introduction

Traditional Friedel-Crafts acylation reactions rely on conventional Lewis acid catalysts like  $\text{AlCl}_3$  or  $\text{FeCl}_3$ , which often require excess quantities and are harmful to the environment. To address these challenges, green chemistry approaches now focus on using renewable plant-based resources for catalyst preparation. *Moringa oleifera* leaves contain natural compounds capable of reducing and stabilizing metal ions, enabling the eco-friendly synthesis of iron particles. These bio-synthesized iron particles can act as efficient, reusable catalysts, reducing the environmental footprint of organic reactions. This work used Moringa leaf extract to prepare  $\text{Fe}_3\text{O}_4$  and zero-valent Fe (ZVI) nanoparticles (NPs) and applied them in Friedel-Crafts acylation reactions, highlighting a sustainable pathway for catalytic organic synthesis.

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## Methodology

### Preparation of Moringa leaf extract

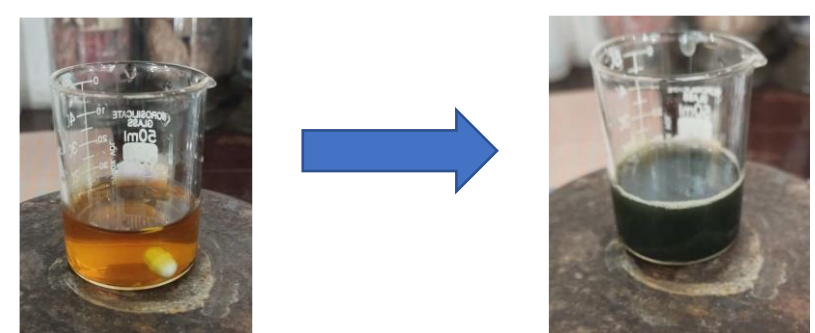


washed, sun-dried, finely ground (10 g), boiled in deionized water (100 ml, 70 °C, 1 h) and filtered

### Synthesis of $\text{Fe}_3\text{O}_4$

#### Plant extract-based synthesis of $\text{Fe}_3\text{O}_4$ NPs (PES- $\text{Fe}_3\text{O}_4$ )<sup>1</sup>

$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  (3.244 g, 20 mmol) and  $(\text{NH}_4)_2\text{SO}_4 \cdot \text{FeSO}_4 \cdot 6\text{H}_2\text{O}$  (3.921 g, 10 mmol) (2:1) mixed with 20 ml of moringa extract, adjusted pH to 11, stirred for 1 h, black precipitate centrifuged, washed, dried at 70 °C for 24 h and stored under  $\text{N}_2$



#### Chemical synthesis of $\text{Fe}_3\text{O}_4$ NPs (CS- $\text{Fe}_3\text{O}_4$ )

Dissolved above salts in degassed water under  $\text{N}_2$ ,  $\text{NaOH}$  (10 ml, 3.0 mol  $\text{dm}^{-3}$ ) added dropwise, black precipitate centrifuged, washed, dried at 70 °C for 24 h and stored under  $\text{N}_2$

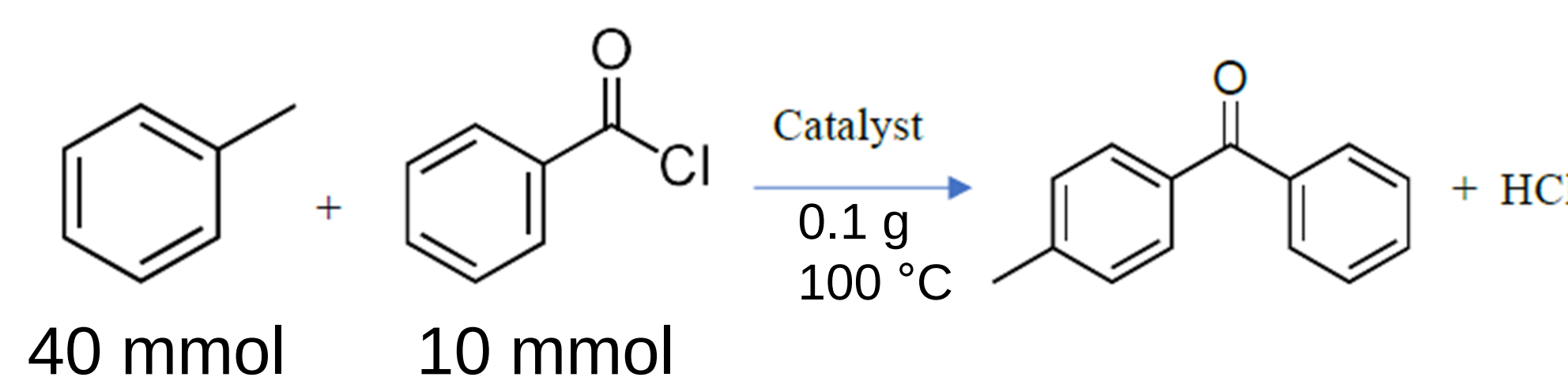
#### Plant extract-based synthesis of ZVI NPs (PES-ZVI)

$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  (10 g, 36 mmol) added to 100 ml of moringa extract, stirred at room temperature for 24 h, product was centrifuged, washed, and vacuum-dried

#### Chemical synthesis of ZVI NPs (CS-ZVI)

$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  (10 g, 36 mmol) dissolved in 100 ml of 30% methanol, pH ~6,  $\text{NaBH}_4$  (50 ml, 3.97 g) added dropwise, stirred for 20 min, precipitate centrifuged, washed, vacuum-dried.

### Benzoylation of toluene<sup>2</sup>



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## Results

### Results of the synthesis of NPs

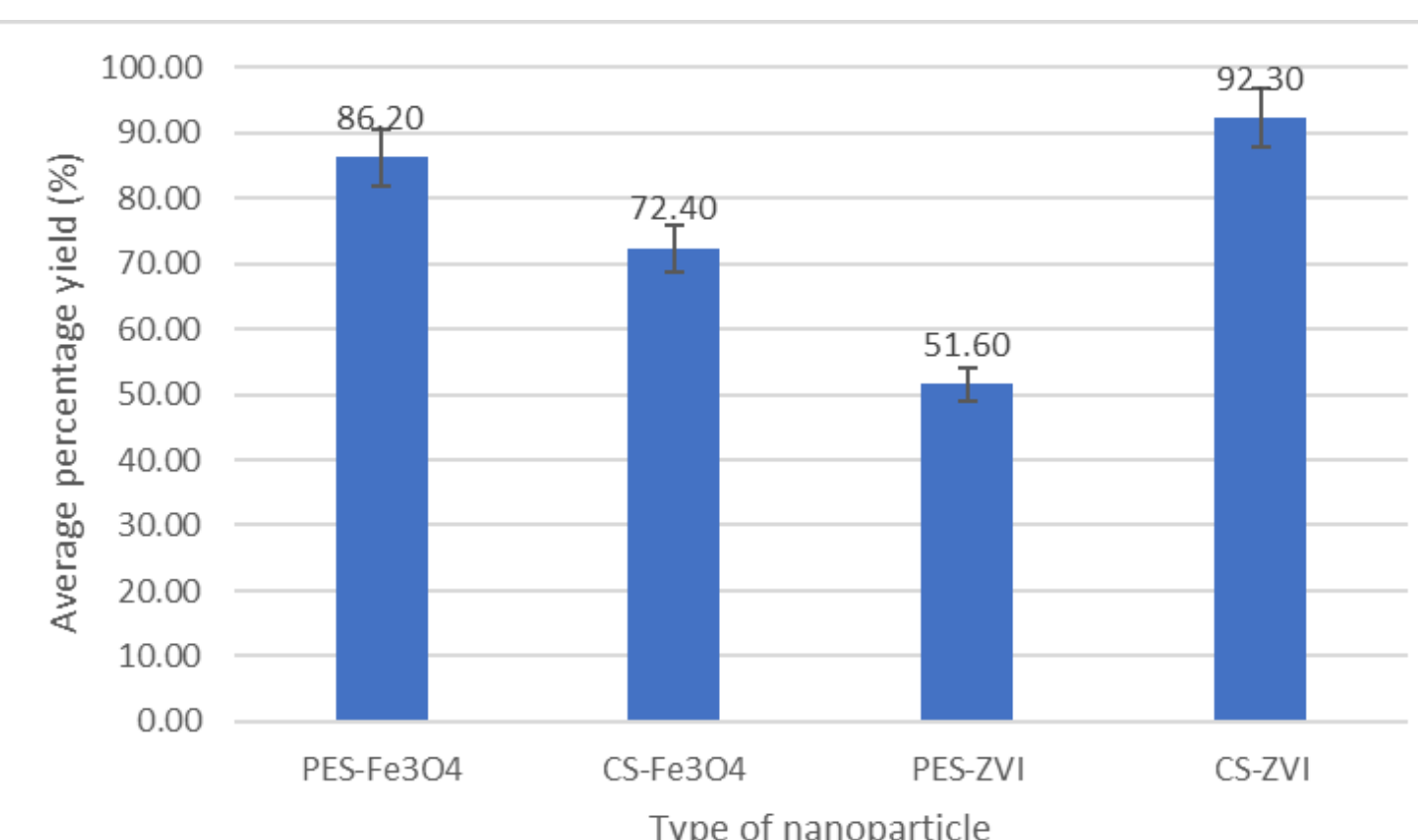


Figure 1: Percentage yields of synthesized NPs



Figure 2: Magnetic properties of NPs

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### Characterization of the synthesized NPs

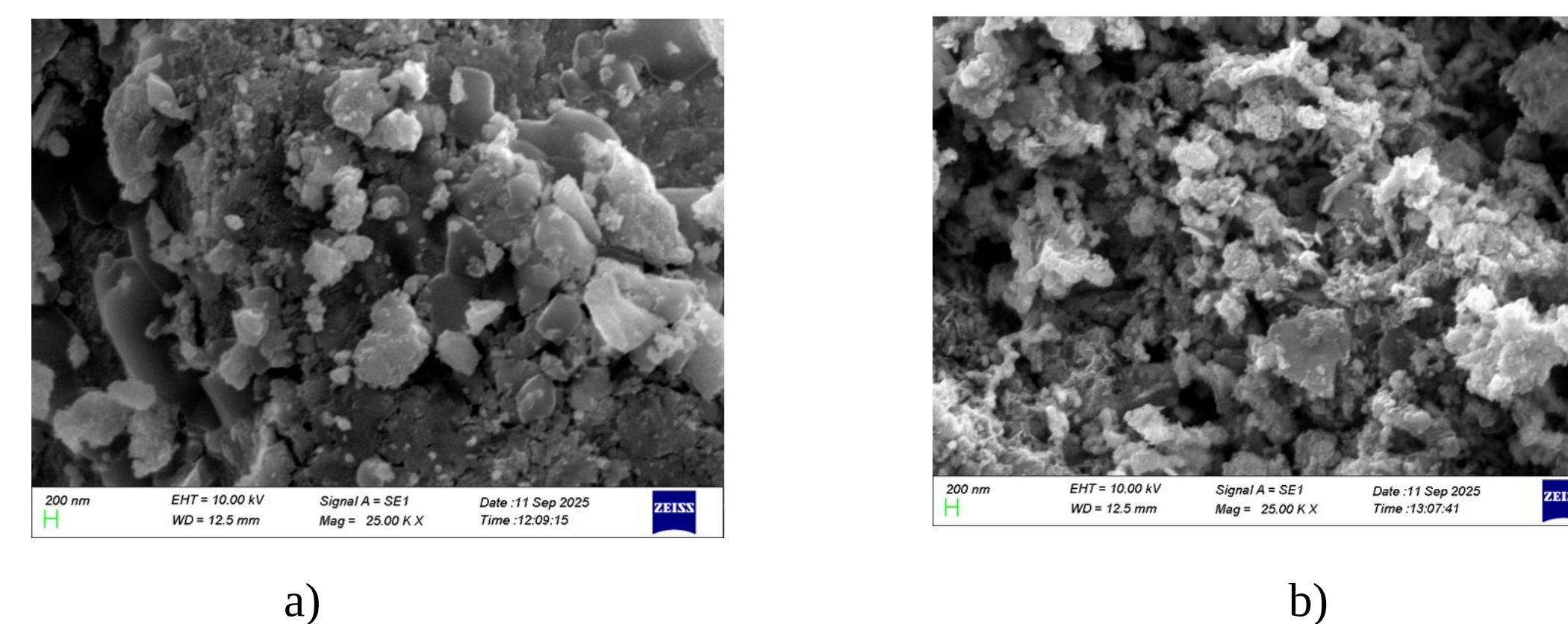


Figure 3: SEM of a) Moringa leaf extract-based  $\text{Fe}_3\text{O}_4$  NPs and b) Moringa leaf extract-based ZVI NPs

Table 1: XRD patterns of a) Moringa leaf extract-based  $\text{Fe}_3\text{O}_4$  NPs and b) Moringa leaf extract-based ZVI NPs

| Type of NP                                                                                   | 2θ values | Phase |
|----------------------------------------------------------------------------------------------|-----------|-------|
| Moringa leaf extract based $\text{Fe}_3\text{O}_4$ NPs                                       | 30.10°    | (220) |
|                                                                                              | 35.42°    | (311) |
|                                                                                              | 43.05°    | (400) |
|                                                                                              | 56.94°    | (511) |
|                                                                                              | 62.51°    | (440) |
| Moringa leaf extract-based ZVI NPs<br>(Characteristic peaks corresponding to $\text{Fe}^0$ ) | 44.6°     | (110) |
|                                                                                              | 65.6°     | (200) |
|                                                                                              | 82.6°     | (211) |

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### Results of the benzoylation of toluene

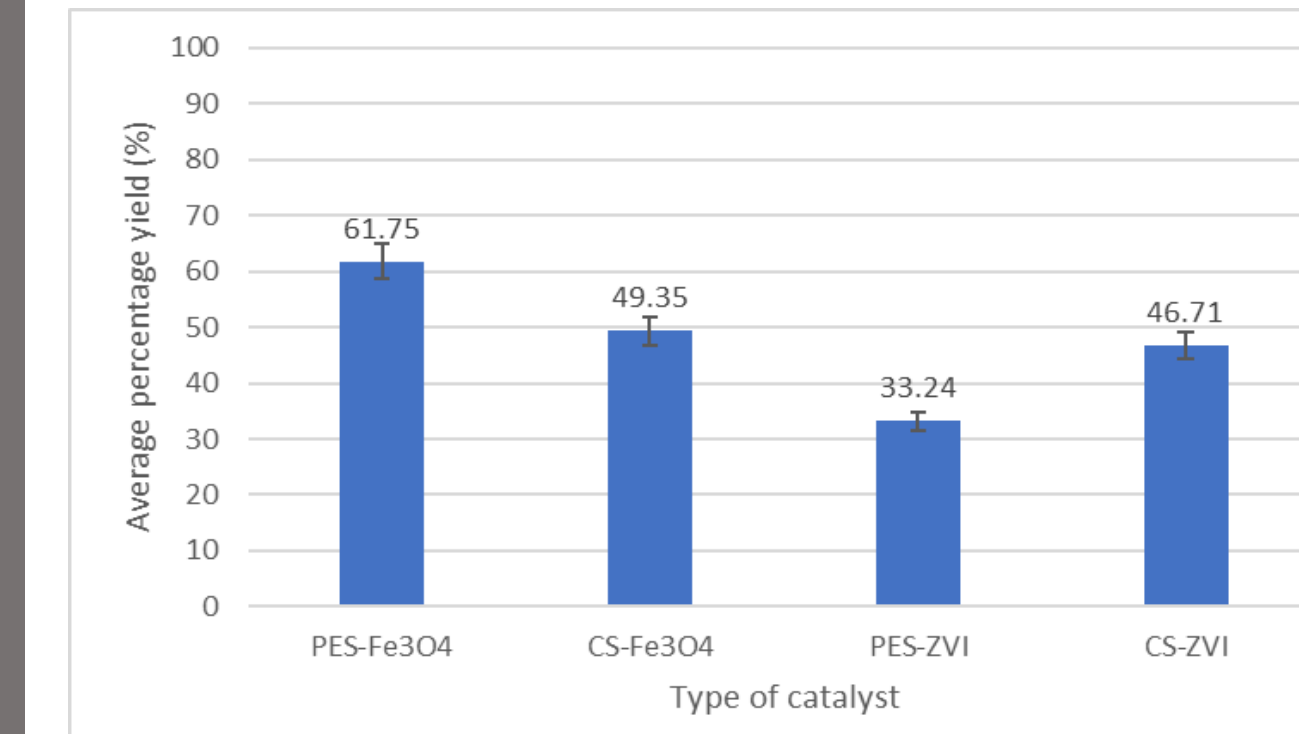


Figure 4: The average percentage yields of the Friedel-Crafts acylation for different catalysts

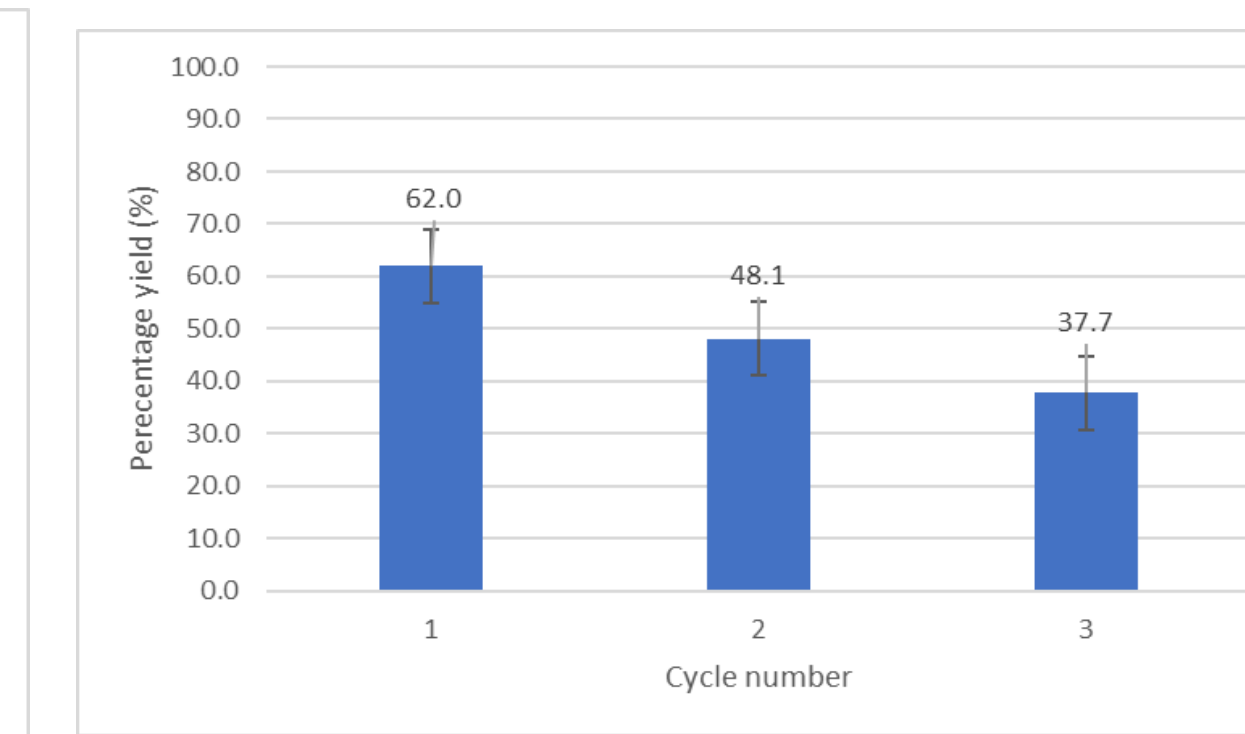


Figure 5: Reusability of Moringa leaf extract-based  $\text{Fe}_3\text{O}_4$  NPs as catalysts

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### Characterization of the product

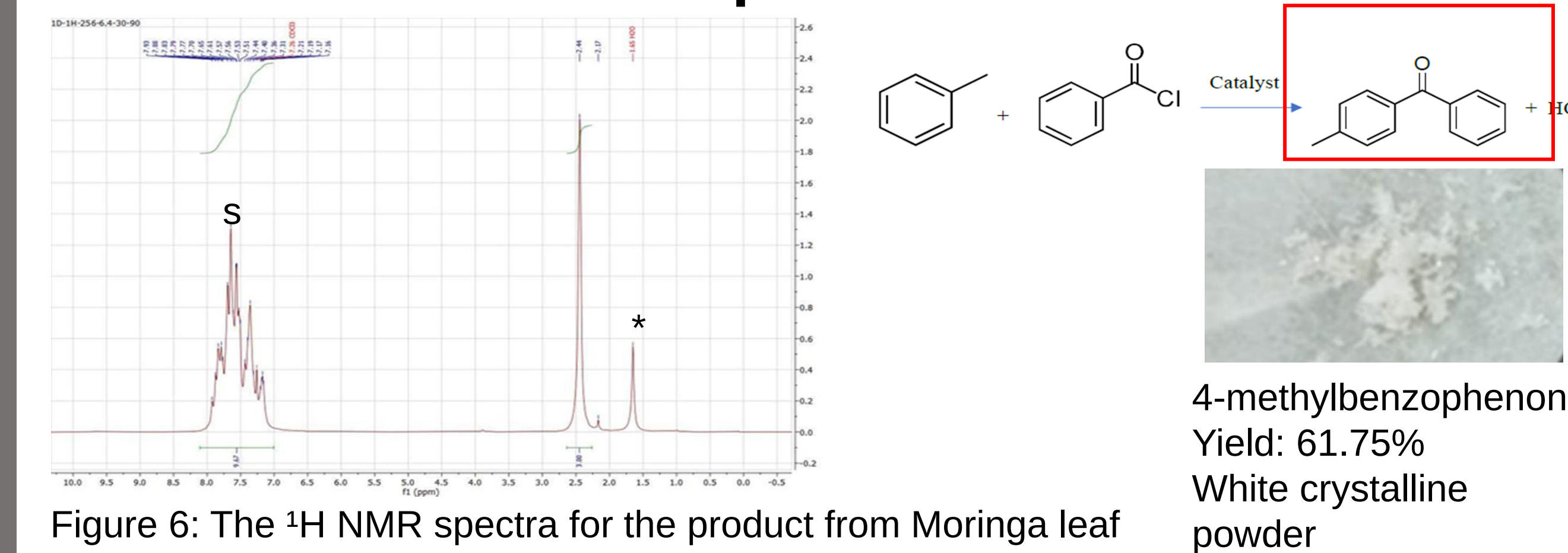


Figure 6: The  $^1\text{H}$  NMR spectra for the product from Moringa leaf extract-based  $\text{Fe}_3\text{O}_4$  NPs as catalyst

\* - water proton s - solvent peak  
 $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  2.43 (s, 3H), 7.14–7.91 (m, 9H)

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## Conclusion

- Moringa leaf extract enabled the plant extract-based synthesis of  $\text{Fe}_3\text{O}_4$  and ZVI NPs.
- Plant-derived  $\text{Fe}_3\text{O}_4$  gave the highest catalytic yield in Friedel-Crafts acylation (61.75%).
- Phytochemicals in the extract acted as natural stabilizers, enhancing activity and stability.
- Demonstrates a sustainable, reusable nano catalyst as an eco-friendly alternative to conventional Lewis acids.

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## References

- Kiwumulo, H. F.; Muwonge, H.; Ibingira, C.; Lubwama, M.; Kirabira, J. B.; Ssekitoileko, R. T., *BMC Research Notes* **2022**, 15 (1).
- Hoseini, S. J.; Nasrabadi, H.; Azizi, M.; Beni, A. S.; Khalifeh, R., **2013**, 43 (12), 1683–1691.

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