Advanced Biorefining with Ionic Liquids – Unlocking Sustainable Materials from Mixed Biomass

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Background and Introduction

Plant-derived fuels and materials synthesised from *cellulose, hemicellulose* and *lignin* found in *lignocellulosic biomass* are a promising alternative to fossil fuels. They can be accessed through various biorefinery processes (Kraft Pulping, pyrolysis, Organosolv etc.). Phase I and Phase II biorefinery is heavily commercialised.

ionoSolv fractionation is a sustainable biorefinery approach to accessing high quality cellulose, lignin and hemicellulose using recyclable protic ionic liquids. ionoSolv is highly successful with single feedstock systems but **mixed biomass feedstocks unexplored.**

Lignocellulosic biomass types:
1. *Woody: softwood (pinewood), hardwood (willow)*
2. *Grassy: straw, grasses (Miscanthus)*

This study aims to contributes to UN Sustainable Development Goals 9,12 and 13.

Biorefinery	Feedstock	Product
Phase I	Single	Single
Phase II	Single	Multiple
Phase III	Multiple	Multiple

Table 1. Biorefinery phases. Adapted from ref. **2,** under terms of CC by 4.0 license https://creativecommons.org/licenses/by/4.0/

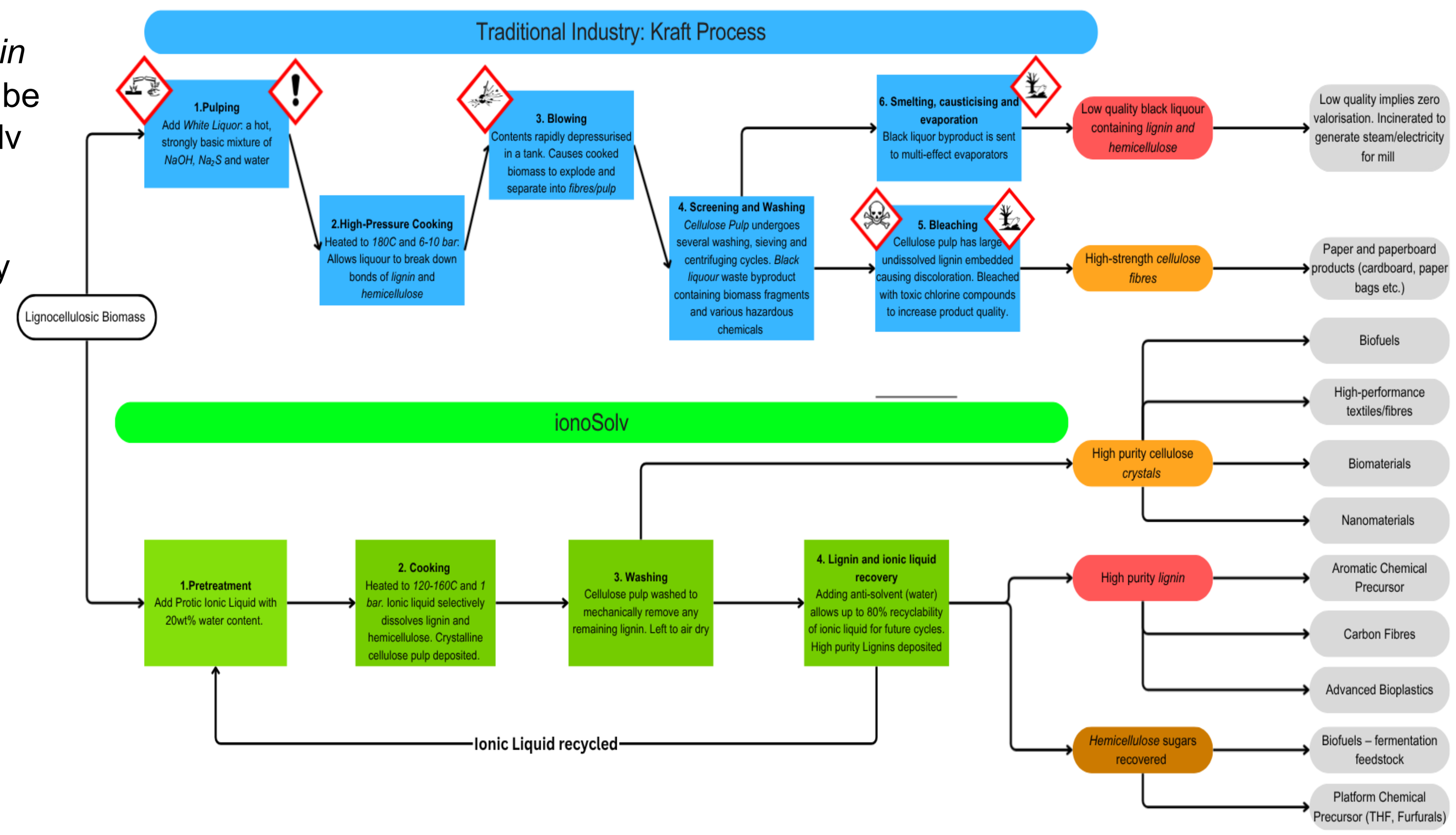


Figure 1. Schematic for Kraft Pulping v ionoSolv. Adapted from ref.**1,** under the terms of CC by 4.0 license https://creativecommons.org/licenses/by/4.0/

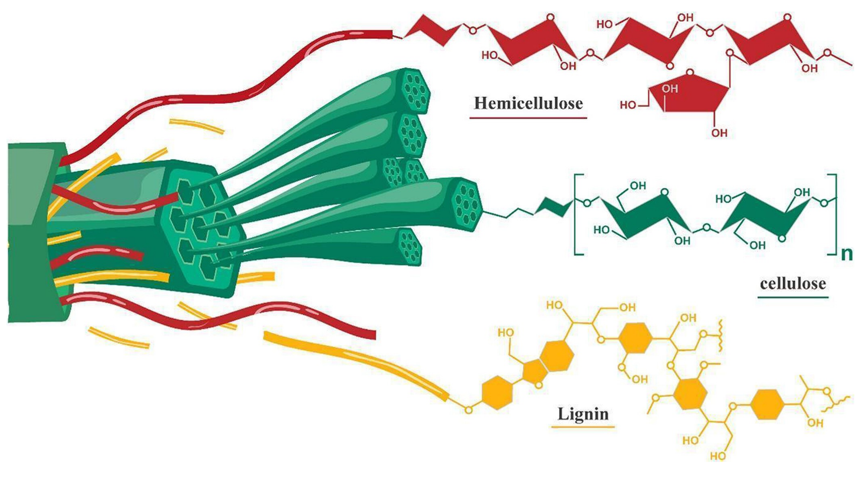
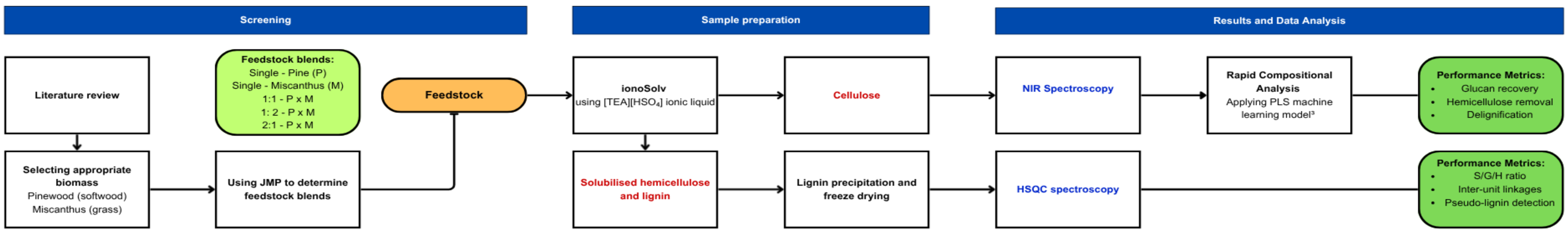


Figure 2. Lignocellulosic biomass composition. Adapted from ref. **1**, under terms of CC by 4.0 license https://creativecommons.org/licenses/by/4.0/

Materials and Methods



Results and Discussion

Compositional Analysis

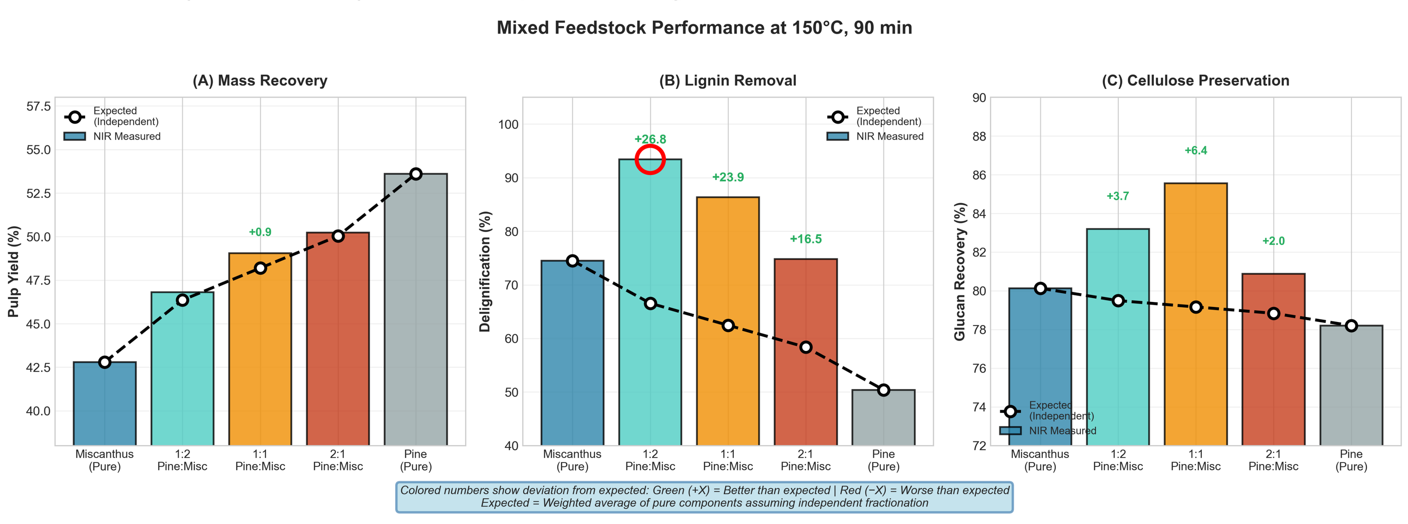
Rapid screening of ionoSolv performance conducted using Near-infrared (NIR) spectroscopy-based compositional analysis

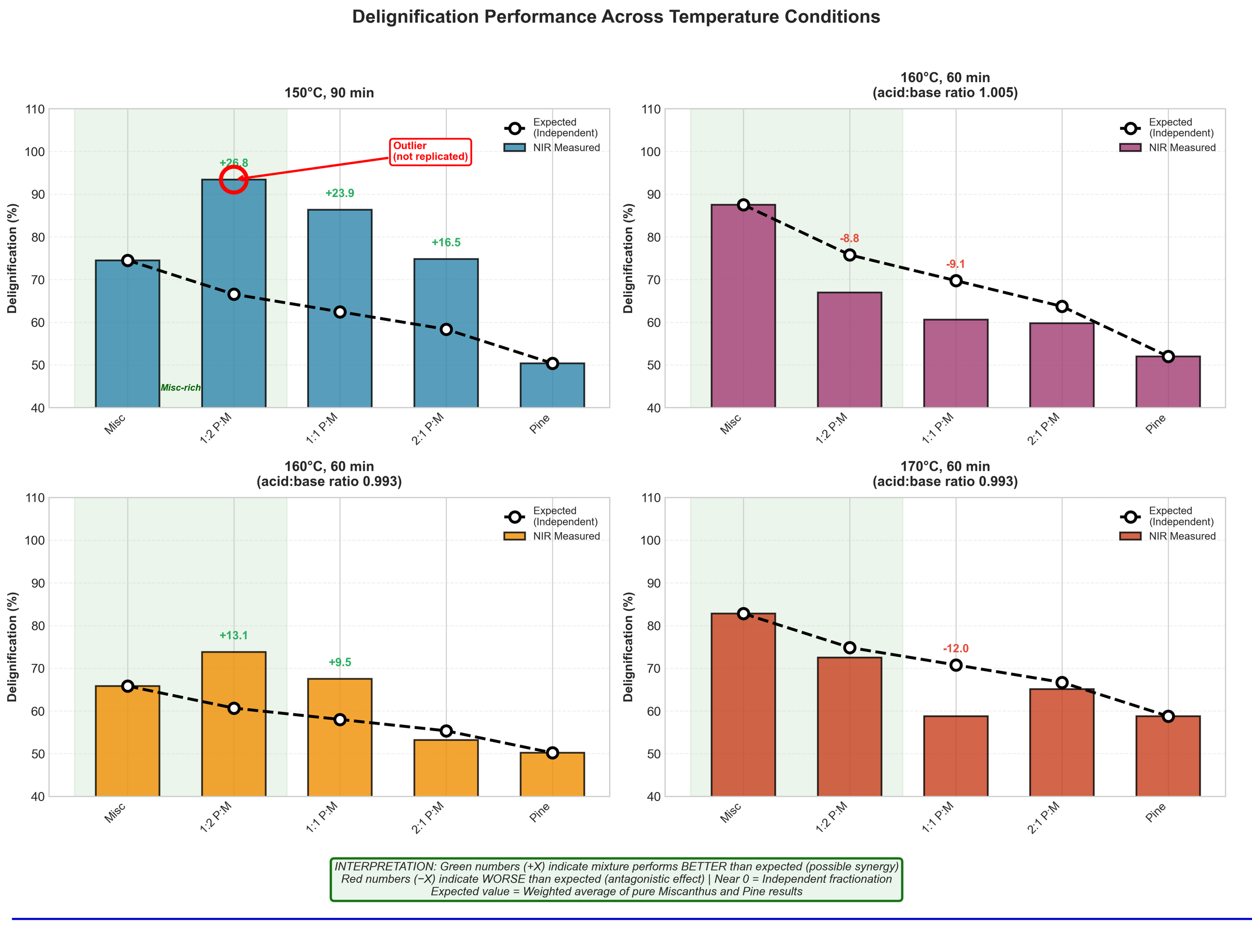
Feedstocks have incompatible optimal processing conditions. Miscanthus achieves significantly higher delignification under identical conditions to pine. Pine is inherently recalcitrant.

Mixed feedstocks displayed intermediate performance generally consistent with weighted averages of pure component behaviour. Miscanthus-rich mixtures (≤33% Pine) achieving 67-75% delignification.

One measurement suggested enhanced performance for the 1:2 P x M ratio (93% delignification), this was not consistently observed across other temperature conditions (67-74% at other temperatures), suggesting measurement variability rather than genuine synergistic effect.

Extreme conditions (170°C, high A-B ratio) caused severe degradation of both components with pseudo-lignin formation. Confirms need for Miscanthus-optimized conditions (150-160°C) when co-processing these feedstocks.





HSQC

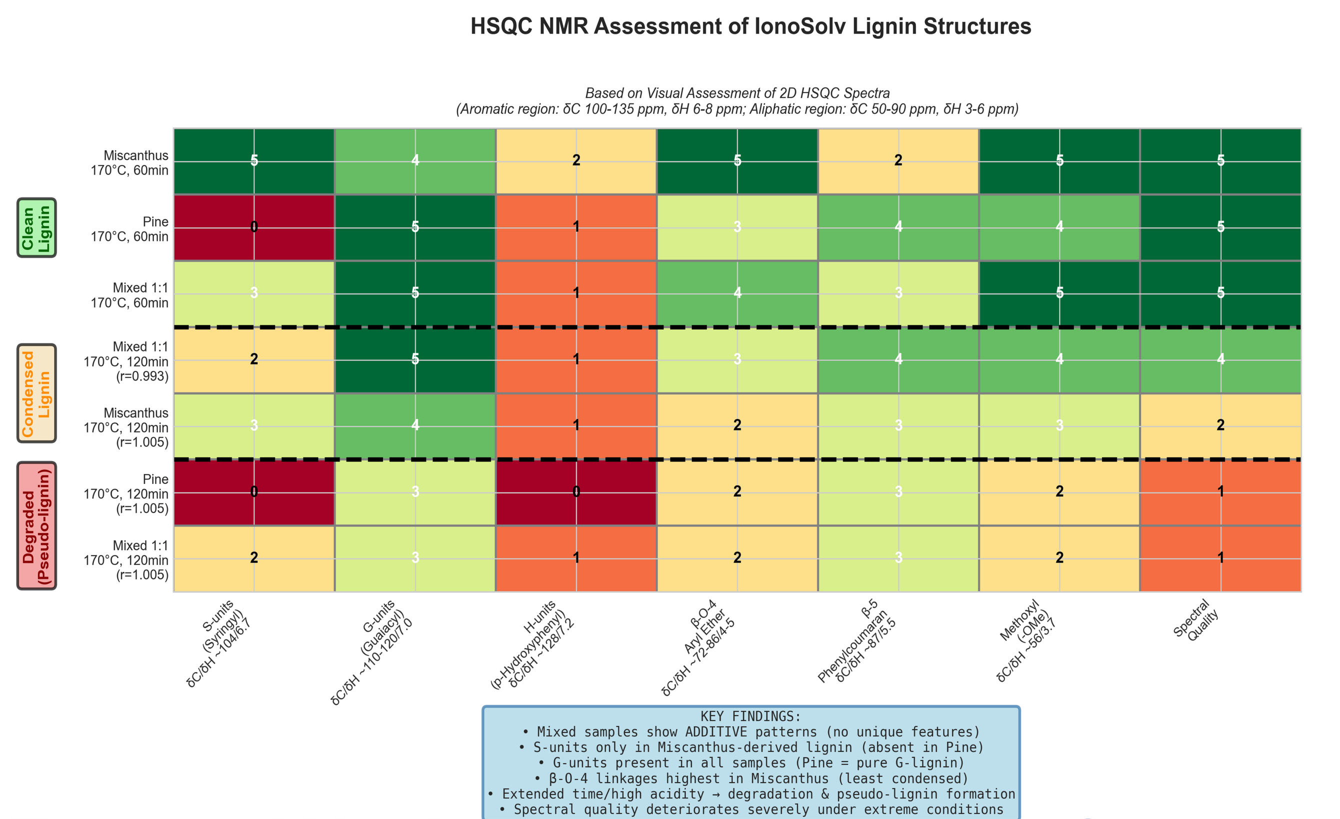
Miscanthus lignin rich in S and G units; Pine exclusively G-units; *mixed samples show both without novel cross-peaks*

Absence of unique S-G linkage signals indicates independent lignin populations. *No cross-condensation*

β-O-4 aryl ether linkages abundant in Miscanthus, reduced in Pine and mixtures

Extended time (120 min) increases condensation and higher acidity (r=1.005) causes severe pseudo-lignin formation

Mixed feedstock lignin consists of two independent populations with no detectable cross-condensation, consistent with sequential extraction kinetics predicted from fundamental lignin chemistry



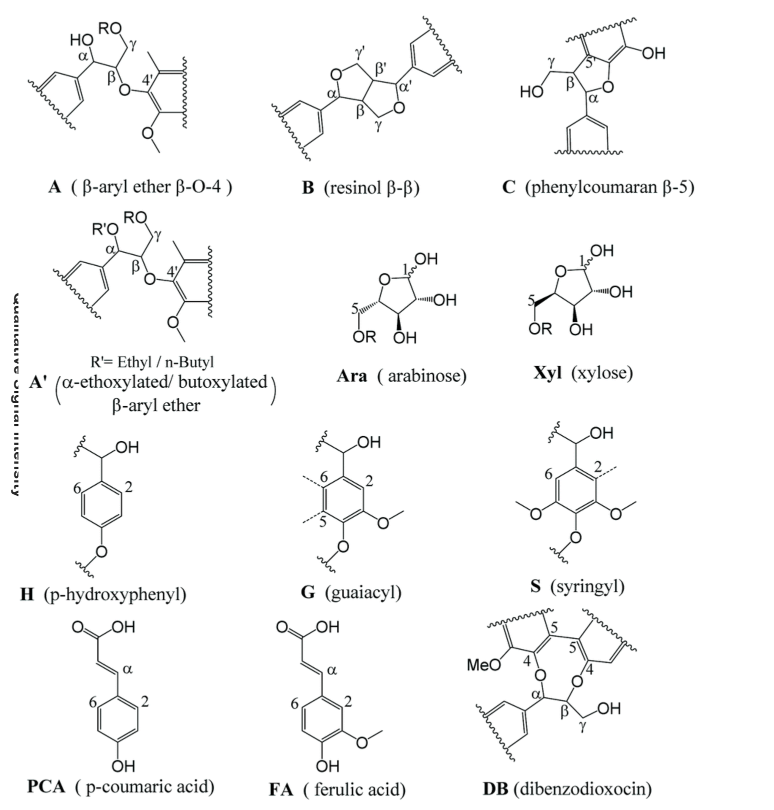


Figure 3. Key lignin substructures and monolignols

Future Work

- do **n ≥ 3** repeats to allow statistical reliability and analysis
- Investigate rheology and molecular dynamics
- Scaling studies: continuous stirred tank reactor, mass transfer effect
- Further NMR experiments
- Techno-economic analysis against single-feedstock biorefinery

Conclusions

Mixed feedstocks is additive with no measured synergy/antagonism

Miscanthus-optimized conditions (150-160°C) needed in pine-containing feedstock blends for favourable yields and delignification

Absence of cross-condensation reinforces that miscanthus and pine extract independently in ionoSolv. Supports two-stage valorisation strategies where Miscanthus-derived S-lignin (higher value, more β-O-4 linkages) can be segregated from Pine-derived G-lignin (more condensed, lower value).

References

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Acknowledgements

The author would like to acknowledge his supervisors and the Hallett research group for their extensive and continued support in this project. The author also acknowledges the Laidlaw Foundation for sponsoring this project.

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