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Rapid assessment of wood chemical properties and pulp yield of *Eucalyptus camaldulensis* in Thailand tree plantations by near infrared spectroscopy for improving wood selection for high quality pulp

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Abstract Near-infrared (NIR) spectroscopy has been demonstrated as a means for rapid nondestructive determination of the chemical composition and final pulp yield of *Eucalyptus camaldulensis* in Thailand tree plantations. Multiple linear regression (MLR) analysis and partial least squares (PLS) analysis were introduced to develop statistical models in terms of calibration equations for total pulp yield, screened pulp yield, and contents of α-cellulose, pentosans, and lignin in wood. In MLR analysis, a reasonably good calibration equation was found only for pentosans (standard error of prediction (SEP): 0.98%). The PLS analysis improved the accuracy of prediction for every criterion variable, especially for pentosans (SEP: 0.91%) and lignin (SEP: 0.52%). Also, in the case of screened pulp yield, we were able to use such a statistical result as an indicator of the characteristics of the pulp and paper. Thus, NIR spectroscopy could be satisfactorily used as an effective assessment technique for *Eucalyptus camaldulensis* plantation trees.

Key words Near infrared spectroscopy · Chemical property · Pulp yield · *Eucalyptus camaldulensis* · Chemometrics

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Introduction

The pulp and paper industry has played an important role in the economic development of Thailand with production expanding year by year. *Eucalyptus camaldulensis* is utilized as the main raw material even at ages of 3–5 years in Thailand because of the high growth rate. In order to improve the productivity of pulp and paper mills, the papermaking industry requires the supply of wood chips of stable quality in terms of lignin and cellulose content, wood density, length of fibre, etc. However, this is difficult to achieve through tree-breeding programs, because wood is a natural resource. The chemical properties and inherent physical properties in pulp and paper of *Eucalyptus* have especially wide varieties, which depend on the plantation site, tree age, and individual specimens.[1]

Recently, near-infrared (NIR) spectroscopy has been recognized as one of the most powerful non-destructive techniques for wood quality analysis.[2–7] Several researchers have examined and proved the ability of NIR spectroscopy to analyze wood chemical properties as well as the inherent physical properties of pulp and paper. Concerning *Eucalyptus* woods, the origins of absorption bands in the NIR spectra of *Eucalyptus globulus* were introduced by Michell and Schimleck.[8] They also demonstrated quantitative analysis of *E. globulus* and *Eucalyptus nitens* plantation woods by NIR spectroscopy.[9] More precise results were obtained for *E. globulus* than for *E. nitens*. Furthermore, *Eucalyptus* tree classification by principal component analysis,[10] estimation of density,[11] or growth strains[12] have also been examined, by which NIR spectroscopy was conclusively identified as a useful technique for pulp and paper industries.

The purpose of this study was to investigate the feasibility of predicting chemical properties and pulp yield using NIR spectroscopy as a selection criterion in the tree breeding and high-quality pulp wood selection programs of *E. camaldulensis* tree plantations in Thailand. This is the first application of NIR techniques to *E. camaldulensis*, although it may not be ideal because of the dark red color of the timber. In this study, the multiple linear regression

(MLR) analysis, in comparison with the partial least squares (PLS) analysis, was introduced to develop statistical models in terms of calibration equation for each trait. Because the chemical and pulp and paper characteristics of *Eucalyptus* sp. depend on location, this research may provide fundamental information related to the silviculture of *E. camaldulensis* in Thailand.

Material and methods

Sample

Wood chips of *Eucalyptus camaldulensis* (tree age: 4–5 years, site: Kanchanaburi Province, Thailand) were supplied by Siam Pulp and Paper (Ratchaburi, Thailand). The samples were separated into two parts, one part for soda pulping and the other part for wet-chemical and NIR analysis. Air-dried wood chips were ground to pass through a 40-mesh screen. Samples retained on a 60-mesh screen were collected for wet-chemical analysis and NIR analysis.

Wet-chemical analysis and pulp yield of wood

Quantitative wet-chemical analyses were performed for lignin, α-cellulose, and pentosans based on standard TAPPI methods.[13] The total and screened pulp yields for soda pulping were also determined for 20-Kappa pulps, which were provided by Siam Pulp and Paper.

NIR analysis

A total of 55 samples of wood meal were prepared for NIR analysis. Each sample was kept at 25°C prior to the NIR measurement. The NIR diffusely reflected spectra were obtained by scanning 20 g of each sample in a standard closed cup using an IA500 spectrophotometer (Bran + Luebbe, Norderstedt, Germany). A total of 36 scans were collected and averaged over the wavelength range from 1100 to 2500 nm at stepwise intervals of 2 nm. Experiments were conducted in triplicate.

Chemometric technique

The 55 samples were divided into two sets of similarly distributed traits. The calibration set, having a wider range of traits, was used to develop the calibration model by means of MLR and PLS analysis. The statistical model was subsequently validated with the second sample set. Raw spectral files were imported into SESAMI (v. 2.00; Bran + Luebbe) and Pirouette (v. 3.02; Infometrix, Woodinville, USA) for data analysis. Quantitative techniques investigated in this work were MLR (SESAMI) and PLS (Pirouette).

The use of MLR in NIR calibration may owe something to its relationship with the traditional spectroscopic method of simultaneous equations for coping with overlapping peaks. The solution to the equation has a form like a linear combination of spectral data. In this study, selection of wavelengths in calibration equations was performed by computer, where all the wavelengths were tried to find the best combination of spectral data as explanatory variables. Thus, they were not chemically but rather statistically selected. The number of wavelengths was varied from two to six to find best calibration equation.

In PLS analyses, the predicted residual error sum-of-squares was calculated for independent validation samples to choose the calibration equation that provided the best results. The wavelength range for PLS analysis was suitably selected to avoid the absorption bands of water (e.g., 1400–1600 nm or 1800–2000 nm).

A number of pretreatment combinations were empirically selected to obtain the spectra prior to calibration which could be best described by the developed model. In this study, multiplicative scattering correction (MSC),[14] smoothing, normalization, first-order and second-order derivations were employed as the spectrum pretreatment algorithms. In this study, we aimed to find a useful calibration equation where the standard error of prediction (SEP) was within 10% in the variation of the data range.

Results and discussion

MLR analysis

Table 1 shows the optimum statistical parameter for each trait by MLR analysis. Thus, pretreatment of NIR spectra and selected wavelengths varied with trait. Figure 1 shows the variation of first derivative NIR spectra with total pulp yield. Figure 2 shows the variation of second derivative NIR spectra with lignin content in *Eucalyptus camalduensis*. It is observed from these figures that there are slight differences between spectra whereas criterion variables (i.e., total pulp yield or lignin content) varied to a degree. The circled wavelengths in Fig. 2 indicate the selected explanatory variables for MLR analysis, and are directly related to the absorption band in the wood sample.[15]

As shown in Table 1, we could not find good calibration equations for required traits except for pentosans. It is clear that the explanatory variables are not necessarily selected from the absorption bands assigned to chemical components in wood because they were not chemically but rather statistically selected. In the case of our analysis, MLR may not be acceptable for making robust calibration equations.

PLS analysis

Table 2 shows the optimum statistical parameter for each trait by PLS analysis. Pretreatment of NIR spectra and selected wavelength ranges varied with trait. PLS analysis improved the accuracy of prediction for every criterion variable. In particular the prediction for pentosans and lignin content satisfied our requirement (SEP was within 10%). Also in the case of screened pulp yield, we were able to

Table 1. Statistical results for multiple linear regression (MLR) analysis

Traits	Data range (%)	Pretreatment	Selected wavelengths (nm)	Calibration		Prediction		
				SEC (%)	MCC	SEP (%)	*r*	Bias (%)[a]
Total pulp yield	42.15–50.40	Sm + 1De	1502, 1607, 2316	1.31	0.78	1.38	0.63	0.17
Screened pulp yield	41.00–49.85	Sm + No + 2De	1148, 1244, 1482[b], 1530, 2500	1.13	0.88	1.65	0.62	0.19
α-Cellulose content	35.51–41.45	Sm + No + 1De	1302, 1758, 2166	0.81	0.84	1.45	0.52	−0.21
Pentosans content	12.60–22.74	Sm + 2De	1802, 1920[b], 2204, 2276[b]	0.60	0.98	0.98	0.90	−0.11
Lignin content	24.65–29.94	Sm + 2De	1688, 2180, 2328[b]	0.71	0.80	0.89	0.63	0.16

MCC, Multiple correlation coefficient; SEC, standard error of calibration; SEP, standard error of prediction; *r*, correlation coefficient between conventionally measured values and NIR predicted values in prediction set; Sm, smoothing (n = 11); No, normalization; 1De, first derivation (n = 15); 2De, second derivation (n = 15)
[a]The value of the intercept constant
[b]Wavelength related to absorption band of wood sample

Table 2. Statistical results for partial least square (PLS) analysis

Traits	Data range (%)	Pretreatment	Used wavelengths (nm)	Calibration			Prediction		
				F_{num}	SEC (%)	*R*	SEP (%)	*r*	Bias (%)[a]
Total pulp yield	42.15–50.40	MSC + 1De	2000–2500	4	1.25	0.82	1.35	0.70	−0.08
Screened pulp yield	41.00–49.85	Sm + MSC + 1De	1600–1800 + 2000–2500	4	1.44	0.91	1.17	0.81	−0.06
α-Cellulose content	35.51–41.45	Sm + MSC + 1De	1600–1800	3	0.92	0.88	1.06	0.63	1.06
Pentosans content	12.60–22.74	Sm + MSC + 2De	1600–1800 + 2000–2500	6	0.57	0.98	0.91	0.94	0.05
Lignin content	24.65–29.94	Sm + MSC + 2De	1600–1800 + 2000–2500	4	0.43	0.92	0.52	0.88	0.12

F_{num}, Number of factors; *R*, correlation coefficient between conventionally measured values and NIR predicted values in calibration set; *r*, correlation coefficient between conventionally measured values and NIR predicted values in prediction set
[a]The value of the intercept constant

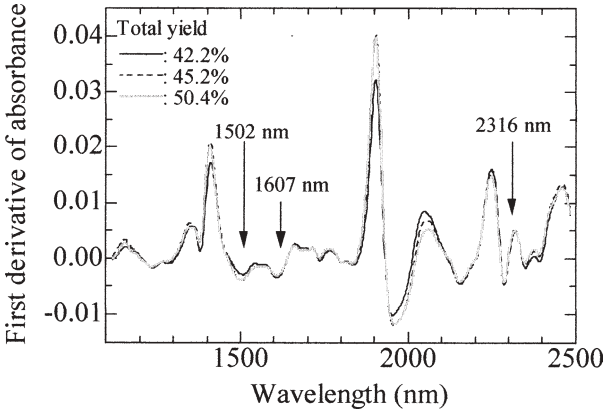


Fig. 1. First derivative NIR spectra of *Eucalyptus camaldulensis* with variation in total pulp yield

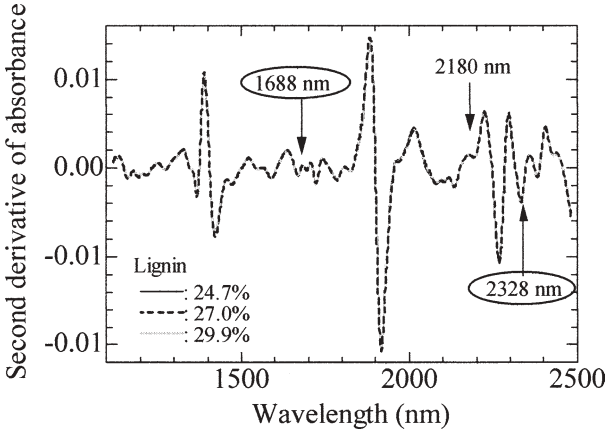


Fig. 2. Second derivative NIR spectra of *Eucalyptus camaldulensis* with variation in lignin content

accept the statistical result as an indicator of the characteristics of the pulp and paper. Figure 3 depicts a spectral loading vector (i.e., solid line) and the first derivative NIR spectrum (i.e., broken line) for total pulp yield. The loading vectors over the absolute value of 0.1, which strongly contribute the PLS analysis, are indicated by arrows. The wavelengths, which are directly assigned to the absorption bands in the wood sample, are also enclosed within circles. In this case, 2276 nm and 2328 nm are assigned to CH_3 and CH in hemicellulose, respectively.[15]

Figure 4 depicts spectral loading vector and second derivative NIR spectrum for lignin content. Wavelength of 1676 nm showing relatively strong loading vector is assigned to CH in aromatic skeletal structure lignin.[8] Therefore, we could identify the spectroscopic and statistical importance for this absorption band. However, the location of the strongest loading vector at 2244 nm was slightly different from the location of the strongest second derivative NIR spectrum at 2276 nm due to CH_3. The PLS analysis, however, is more robust and a better predictor and would be improved with more data.

The accuracy of prediction for each trait is summarized in Figure 5. We could not find a good calibration equation for α-cellulose with MLR or PLS analysis. This point should be considered in future research.

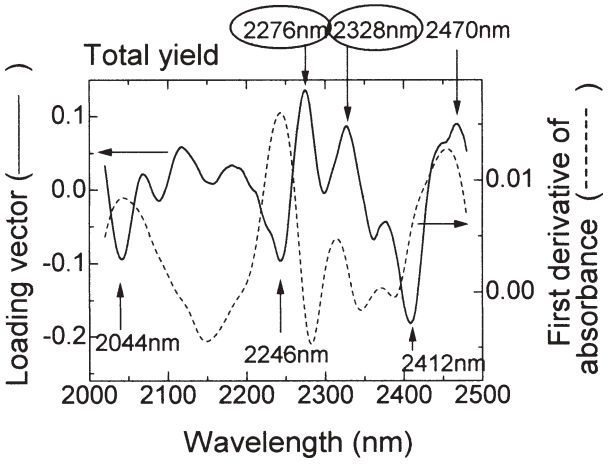


Fig. 3. Spectral loading vector (*solid line*) for partial least squares (PLS) analysis for total pulp yield and first derivative NIR spectrum (*broken line*)

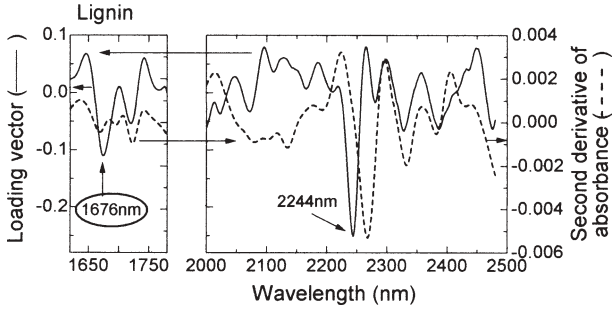
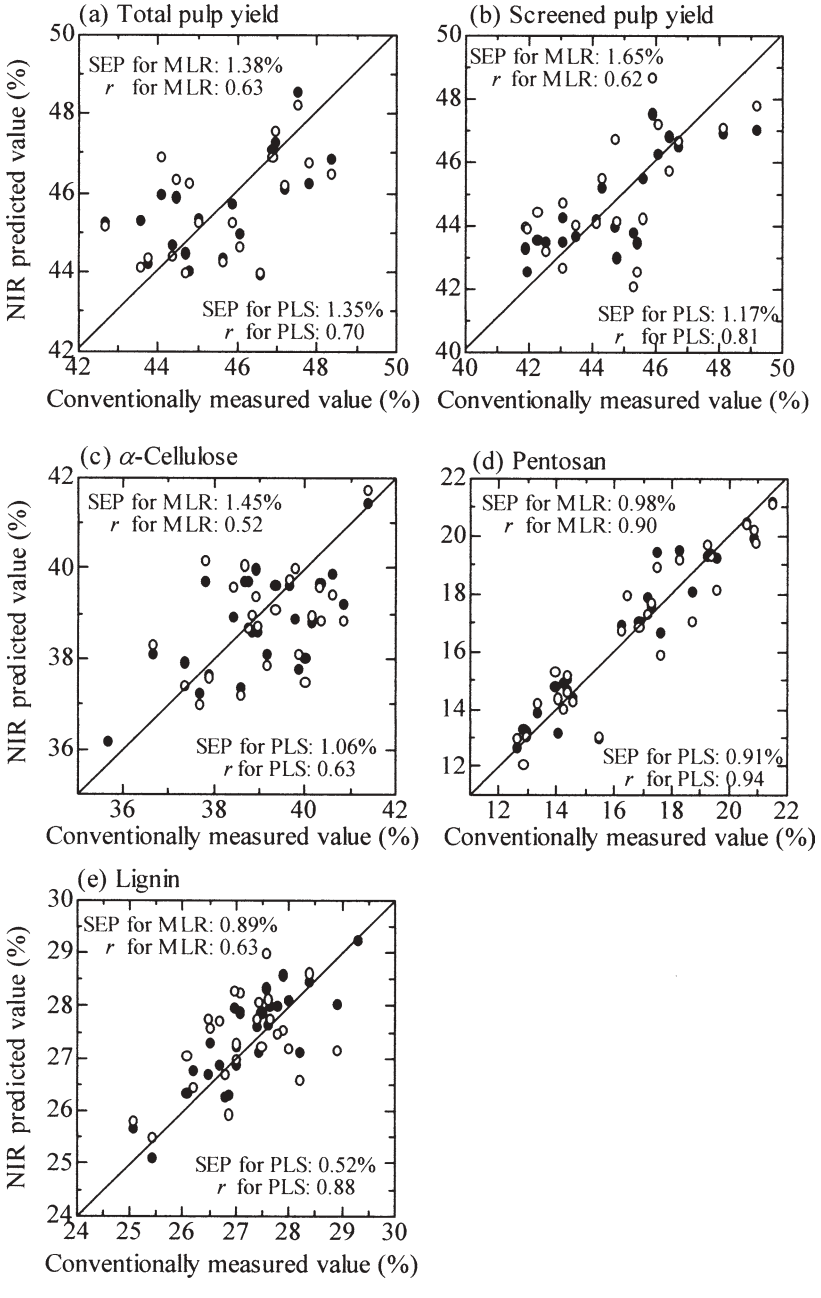


Fig. 4. Spectral loading vector (*solid line*) for PLS analysis for lignin content and second derivative NIR spectrum (*broken line*)

Fig. 5. Prediction of **a** total pulp yield, **b** screened pulp yield, **c** α-cellulose content, **d** pentosans content, and **e** lignin content by multiple linear regression (MLR) calibration (*white circles*) compared with PLS calibration (*black circles*). *SEP*, standard error of prediction, *r*, correlation coefficient between conventionally measured value and actual NIR values



Conclusions

In this study, the feasibility of predicting chemical composition and final pulp yield for *Eucalyptus camaldulensis* woods in Thailand using NIR spectroscopy was examined.

The optimum calibration equations for each constituent were developed with variation of either pretreatment of NIR spectra or selected wavelengths. The MLR analysis indicated that pentosans could be accurately predicted with the calibration equation. PLS analysis improved the accuracy of prediction for every trait. In particular, the predictions for both pentosans and lignin were improved. Also in the case of screen yield of pulp, we were able to accept such statistical results as indicators of the characteristics of pulp and paper properties.

The spectral loading vector for total yield of the pulp showed that some of the vectors strongly contributed to the PLS analysis within the spectral range between 2000 nm and 2500 nm. In the case of lignin, the loading vector at 1676 nm was found as relatively strong information which was assigned to CH in aromatic skeletal structure.

Thus, NIR spectroscopy could be satisfactorily used as a means for rapid evaluation of chemical properties or pulp yield for *Eucalyptus camaldulensis*.

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