DETERMINATION OF RESIN DISTRIBUTION AND COVERAGE IN MDF BY FIBER STAINING

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(Received November 2003)

ABSTRACT

A staining procedure to visualize amino-based resins on MDF fiber has been developed in which dry-blended and blowline-resinated MDF fiber can be stained and resin features on the fiber assessed by confocal microscopy and image analysis. Resin coverage and distribution results determined on stained dry-blended fiber resinated using differing spray atomization regimes were found comparable to those from fluorescently labelled UF resin. Stained blowline-resinated fiber from several MDF mills were analyzed to compare resin distributions. Differences in resin coverage and distribution were observed for changes to resin loading and for modifications to blowline and nozzle parameters. This fiber staining method has the potential to be used as a tool to assess the influence of varying MDF processing conditions for line improvements, quality control, and trouble-shooting applications.

INTRODUCTION

Understanding how differing processing parameters influence the performance of resin on fiber has potential to provide opportunities for improved manufacturing processes of medium density fiberboard (MDF) as well as other wood-based composite panels (Murmanis et al. 1986; Kamke et al. 1996; Loxton et al. 2003). A better utilization of resin may lead to improved panel properties or reduced costs by lowering resin consumption while maintaining panel performance. Reduced resin loading also may potentially lower panel emissions. One key to improving resin performance is the ability to visualize and quantify resin distribution and coverage on fiber in MDF. In MDF panel manufacture, urea formaldehyde (UF) resin is the principal binder (Christjanson et al. 2002). Because of its colorless, opaque nature, this resin cannot be distinguished by conventional light microscopy without enhancement (Donaldson and Lomax 1989). To visualize UF resin, researchers have generally tried either: the inclusion of a dye (Ginzel and Stegman 1970; Albritton et al. 1978; Kamke et al. 2002); label (Loxton et al. 2003; Thumm et al. 2001; Feng and Hutter 2000), or staining (Kamke

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Wood and Fiber Science, 37(2), 2005, pp. 258–269

et al. 2000) in conjunction with various microscopy methods. In the case of our own work, this has included coupling image analysis with either confocal laser scanning microscopy (CSLM) and a chromophore chemically bound to the resin (Loxton et al. 2003), or the inclusion of a UV dye with the resin and fluorescence microscopy to enable the resin to be observed on fiber (Kamke et al. 2002). In each case, these methods have been utilized both in the laboratory and at mill-scale trials to visualize resin and attempt to relate resin distribution to product performance. However successful, in each case, both methods require some pretreatment of the UF resin, as do other reported methods such as using copper salts and electron microscopy (Thumm et al. 2001; Feng and Hutter 2000).

While labelling resin has provided valuable insights into the MDF manufacturing process via resin visualization, ideally a method to characterize UF resin distribution without any pretreatment or mixing of additives would be more versatile. This may enable resin distribution analysis to become routine, or more likely, to be used in quality assurance and trouble-shooting. One of the possibilities to measure the presence of resin on fiber directly is X-ray photon spectroscopy, but the application of this technique has only limited success (Grigsby et al. 2004; Grigsby and Thumm 2004). A technique that shows the most potential as a routine method for resin distribution analysis is one based on applying a stain to resinated fiber (Kamke et al. 2000). In this method, resinated fiber is chemically stained to distinguish the resin component on the fiber. It was envisaged that this staining method would be comparable to using a resin containing a chromophore so that resin distribution analysis could be readily undertaken on resinated fiber sampled directly from a mill, either as a comparative study, quality control, or for trouble-shooting applications. However, when this method was used in a mill, this staining procedure proved unsuccessful, with variable, inconclusive coverage results gained from fluorescence microscopy analysis of stained fiber (Kamke et al. 2000).

The staining method has now been developed further, and reported here are the validation and application of this staining procedure to successfully visualize amino-based resins on dry-blended and blowline-resinated MDF fiber by coupling CLSM with a modified staining procedure. The method was used to examine resinated fiber obtained from various MDF mills, analyzing resin coverage and distribution on fiber sampled at various points in the MDF process. In addition, the effects on resin coverage and distribution by changing common process variables within the mill environment were examined.

METHODS AND MATERIALS

Materials

The dye acriflavine (3,6-diamino-10-methylacridinium chloride) used in this work was purchased from Acros Chemicals and used as received at a concentration of 0.5% (w/w) in deionized water. Formalin solution (35%) was obtained from BDH Chemicals Ltd, and unless stated, was used as received. The fiber used in laboratory dry-blending was obtained by refining radiata pine chips in a pilot plant at 350 kWh/odt with a preheater (172°C) residence time of 3.0 min at 7.5 bar. After exiting the blowline, the fiber was collected and forced air-dried at ambient temperature to a moisture content of *ca.* 10% prior to use. The UF resin used to resinate laboratory fiber was U726, obtained from Orica Adhesives and Resins Ltd (Hornby, New Zealand). Labelled resin was prepared as described in Loxton et al. 2003. Resinated fiber sourced at mills was principally from virgin radiata pine chip furnish and was obtained while running typical MDF production parameters.

Resination and staining of laboratory fiber

Resin was sprayed onto fiber using a twin fluid DeVilbiss spray gun employing coarse, fine, or standard (i.e. medium) spray regimes (Grigsby and Thumm 2004). Generally, resin was sprayed in over 60 s while fiber (500 g) circulated through a 12-m closed loop blender (150 mm dia.) at 18 m/s. The fiber continued to circulate for a further 300 s and then was directed from the loop. Resinated fiber suitable for stain-

ing was sub-sampled from this freshly resinated fiber.

Original staining method.—Dry fiber (0.5 g) was added to 20 mL of a staining solution consisting of 1 part acriflavine solution (0.5%w/w), 1 part 50% HCl acid, and 5 parts water. The solution was gently shaken to immerse all fiber and then allowed to soak. After 3 min, the suspension was filtered and washed with water (approximately 500 mL) until no further dye was washed from the fiber and then air-dried.

Staining of blowline-resinated fiber: Refinement of staining procedure

Resinated fiber was obtained from a commercial MDF mill operation during normal production. The fiber, having an approximate 8% resin loading, was sampled from blowline ports immediately after the resin nozzle injection point (near the refiner) and at the end of the blowline (start-up cyclone), as well as after the fiber had passed through the dryer, prior to forming. Unresinated fiber used as a reference was obtained by suspending resin injection and dumping fiber to the start-up cyclone.

Because of differing inherent moisture contents, the quantity of fiber used in staining was 1.0 g for fiber sampled directly from the blowline and 0.5 g of fiber sampled after the dryer. Unless stated, this fiber was stained within a few minutes after being sampled.

Adapted staining method.—The staining solution consisted of 1 part acriflavine solution (0.5%), 1 part 50% HCl, and 3 parts water. The fiber was weighed out and mixed into 20 mL of staining solution and allowed to soak for 3 min under slight agitation. The fiber was then filtered and immediately immersed in 20 mL of 20% formalin solution. After 2 min this was filtered and washed with water, passing water through the filter pad until no further color was present in the filtrate.

Analysis of resin loading on fiber

Actual fiber resin contents were calculated, using nitrogen elemental analysis performed on a LECO CNS-2000 Analyzer. The % nitrogen results were corrected for moisture content and the % resin loading calculated using Eq. (1):

$$\text{Resin loading} = \left(\frac{\%\text{N}_{\text{panel}}}{\%\text{N}_{\text{resin}}}\right)\times 100 \qquad (1)$$

where: N_{panel} was the % nitrogen of panel; and N_{resin} is % nitrogen of resin.

Confocal laser scanning microscopy (Leica TCS)

The microscopy set up was similar to that used with rhodamine fluorescently labelled UF resin (Loxton et al. 2003). Stained fibers were mounted dry onto glass slides. A reference fiber that was identically stained, but unresinated, was also mounted on the same glass slide. The sample was recorded using two photomultipliers each fitted with a different filter set. The filter settings of the microscope were optimized to ensure maximum distinction between stained resin and fiber. For each fiber sample, at least 10 images, each having an area of 1 mm^2 and consisting of a series of optical slices recorded to a depth of about 80 μm were acquired. These slices were converted into a 2D projection using a maximum intensity projection method. Images were then subjected to image analysis to determine the amount of stained resin present on the fiber and the size of the resin features. Although the filter settings had been optimized, images were recorded with some presence of fiber in the "resin channel." To suppress the fiber presence in the resin channel, a threshold was applied in the image analysis routine to ensure that fiber fluorescence did not contribute to the observed resin coverage. The identically stained unresinated fiber reference was used to determine the correct threshold limit for the coverage analysis.

Fluorescence microscopy

Resinated and unresinated fiber, stained using the adapted method, were analyzed using a Zeiss Axioplan 2 fluorescence microscope fitted with an AxioCam HRc digital camera. The settings em-

ployed used a 20 × 0.4 (200x magnification) lens and filter set consisting of a G365 glass filter for excitation (with maximum light intensity at 365 nm), beam splitter FT 395, and emission LP 420.

RESULTS AND DISCUSSION

Shown in Fig. 1 are CLSM images of stained, resinated fiber and unresinated fiber. In these images, the presence of resin appears as red and the MDF fiber as yellow-green. For fiber stained using the original method, it can be seen, especially in the image of unresinated fiber that staining left "artifacts" on the fiber, which were of similar appearance to stained resin. Additionally, there was a further complication as the images from the CLSM had little contrast between stained fiber and resin, which made image analysis and interpretation difficult. The acriflavine stain used in the procedure is also known as a stain that is used to distinguish lignin from carbohydrate (Donaldson 2002). This could be the origin of the pale orange features on the fibers (Fig. 1). Lack of contrast and artifacts causes difficulties as image analysis requires images to be thresholded, and if the orange color is suppressed, then most resin (appearing as red-orange) will invariably be suppressed also.

Through the application of aminoplast chemistry (Pizzi 1983) and image analysis, it was determined that a higher staining solution concentration, the presence of formaldehyde, and the use of flexible image thresholding were each required for successful staining and analysis of commercial blowline-resinated and laboratory dry-blended fiber by CLSM. This led to a staining procedure (adapted method) in which resin features could be distinguished on resinated fiber by increasing the contrast between resin and fiber and eliminating the appearance of artifacts on unresinated fiber (Fig. 2).

Analysis of blowline-resinated fiber stained using the original method gave no distinction between resinated and unresinated fiber at any image analysis threshold setting (Fig. 3). When formalin solution was introduced as a washing solution for stained fibers, there was an apparent enhancement of the fiber fluorescence creating a greater contrast between the resin component, giving a clear separation of resinated and unresinated fiber in CLSM image thresholding. This enabled some distinction of resin from fiber in image analysis. However, it was the combination of higher stain concentration and the additional washing with formalin solution (adapted method) which led to the largest observed threshold separation of resin and fiber for

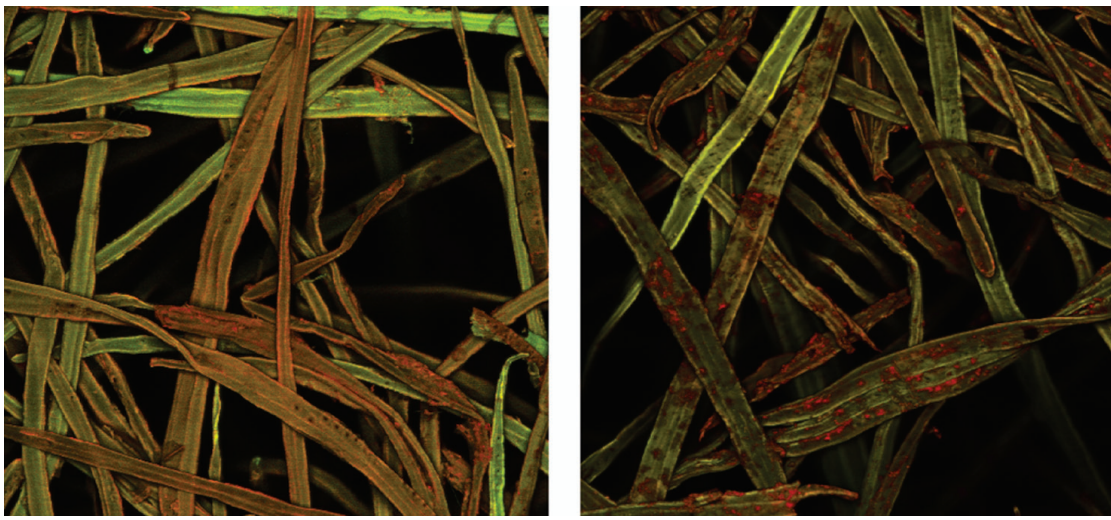

FIG. 1. CLSM images comparing stained unresinated fiber (left) with resinated fiber (right) stained using the original staining method.

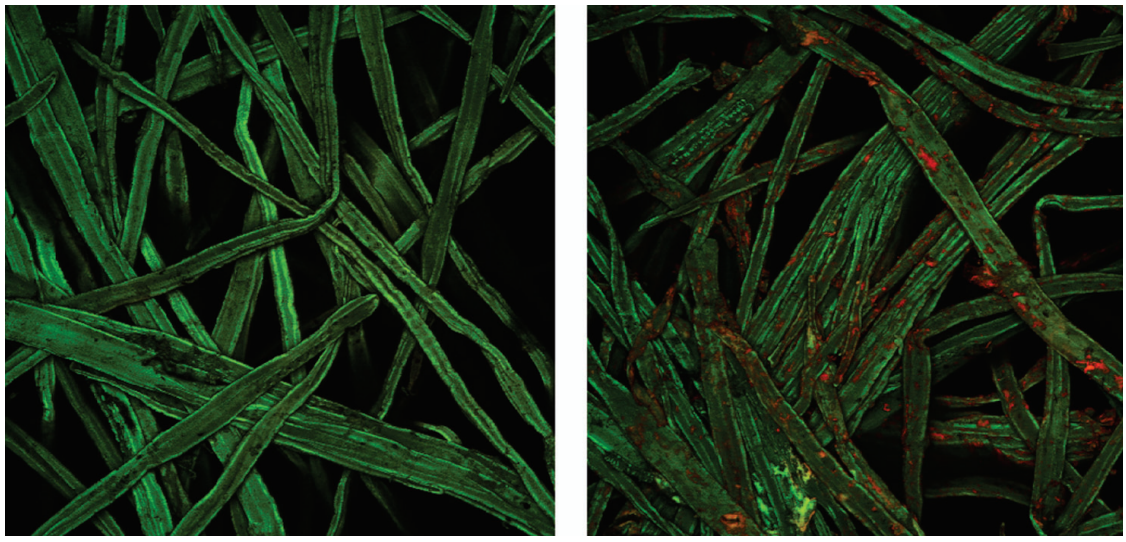

FIG. 2. CLSM images comparing stained unresinated fiber (left) with resinated fiber (right) stained using the adapted staining method.

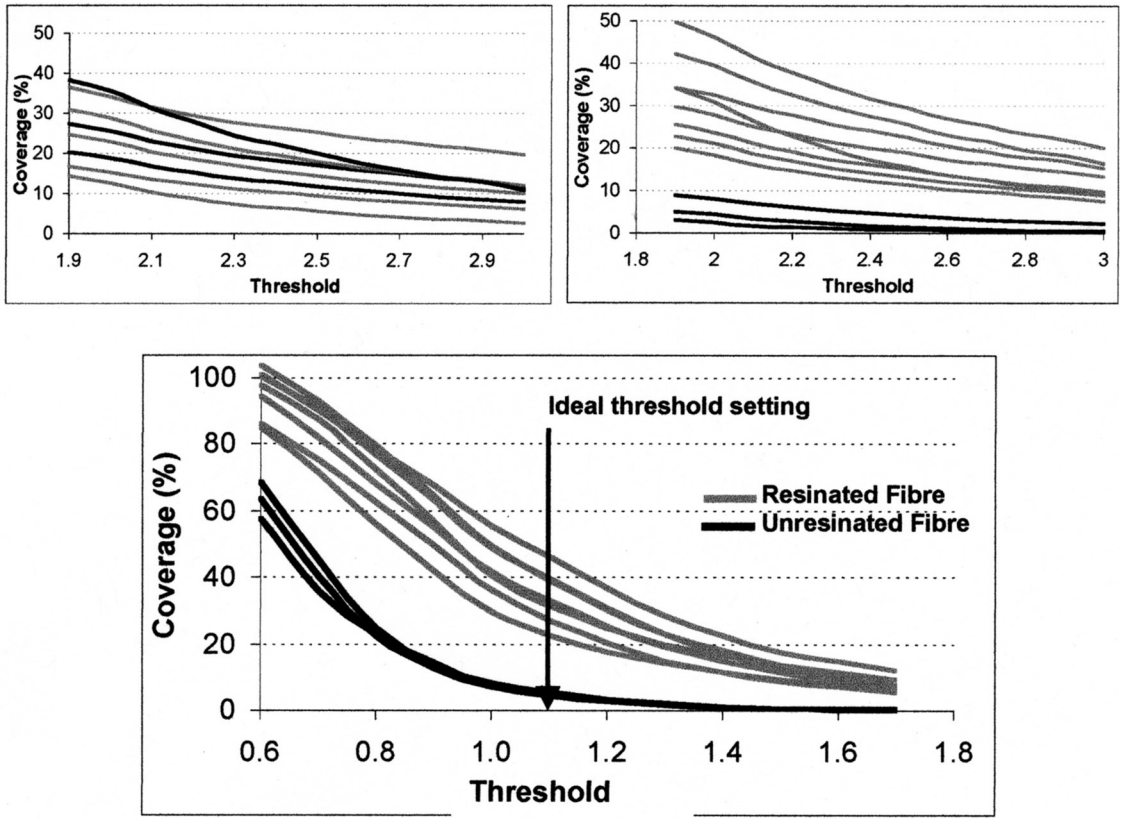

FIG. 3. The effect image thresholding has on resin coverage of stained mill fiber stained using the original method (left), with additional formaldehyde solution (right), and using the adapted method (below).

blowline-resinated fibers. This increased contrast between resin and fiber allowed for lower threshold settings and therefore resin coverage values that were more in line with those previously reported (Kamke et al. 2002).

After adapting the fiber staining procedure for CLSM, work was undertaken on validating the technique. Resin distribution and coverage, obtained from image analysis of dry-blended fiber stained using the adapted method, were compared with those from fluorescently labelled resin and differing resin droplet size spray regimes that had been established in previous work (Grigsby et al. 2004). The results for resin coverage and distribution from stained fiber resinated with differing resin droplet sprays are shown in Figs. 4 and 5, respectively. Figure 5 shows a histogram for the resin droplet size distribution at 12% resin loading. The values for each category are area-weighted, i.e., they show how much each droplet size is contributing to observed coverage (Loxton et al. 2003). This analysis distinguished higher proportions of smaller drops on the fiber formed with a fine resin atomization spray compared to the rela-

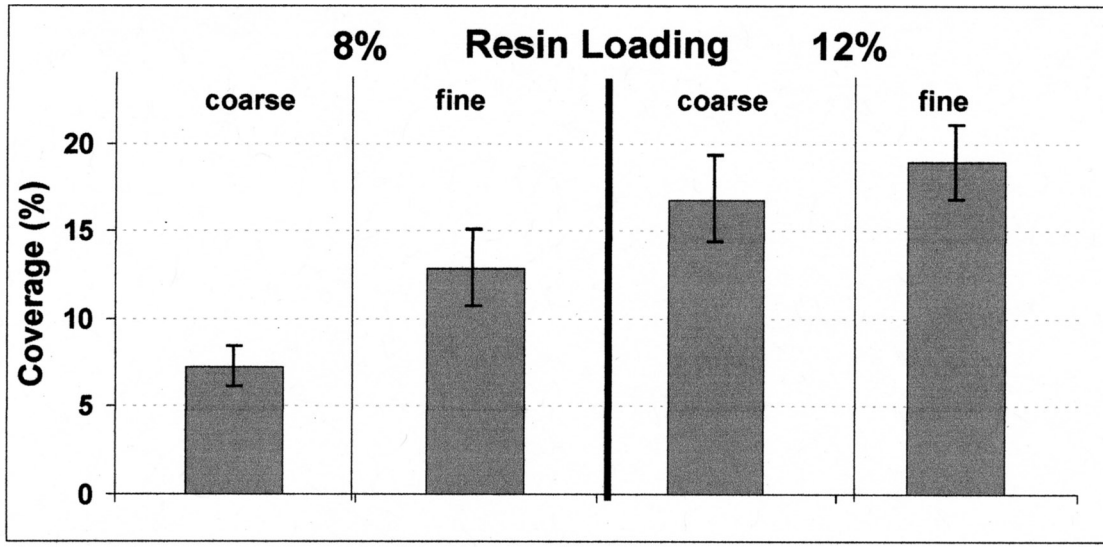

FIG. 4. A comparison of resin coverage of stained dry-blended fiber resinated with coarse and fine spray regimes for 8% and 12% by weight resin loading levels.

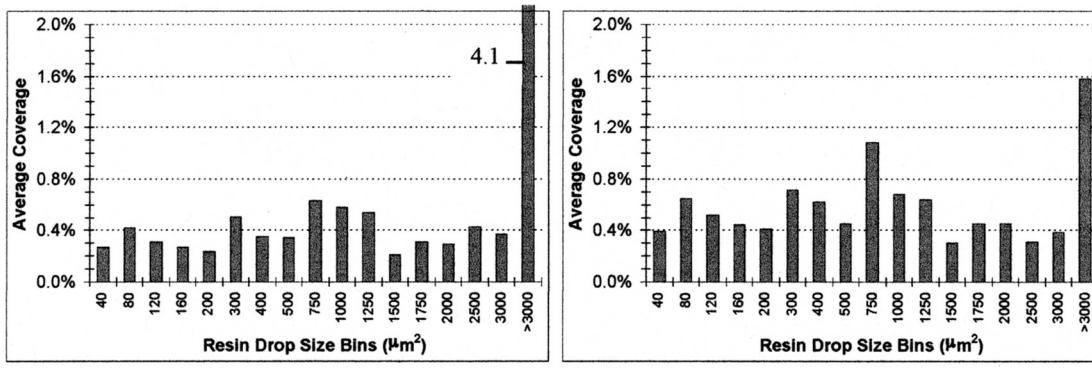

FIG. 5. A comparison of resin distribution results of stained fiber (method 2) resinated with coarse (left) and fine (right) spray regimes.

tively coarse droplet spray. For this coarse spray regime, in which resin was sprayed with sufficient atomization pressure to just avoid resin spotting in panels, the resin distribution had relatively greater proportions of larger droplets (>1000 μm^2) and was dominated by resin features having coverage greater than >3000 μm^2. Image analysis also determined a difference in coverage between the coarse and fine sprays at a resin loading of 8%. No difference was found between the two regimes at higher resin loading (12%). Each of these findings is consistent with those observed using labelled resin (Loxton et al. 2003).

Part of this staining method is the use of a strong acidic medium to ensure the coupling of acriflavine with resin on the fiber, but also to cure and immobilize the resin (Kamke et al. 2000). The low pH, together with the necessity to agitate fibers during staining may cause the resin to be either dislodged or be redistributed on the fiber during the staining process. Fiber was resinated with labelled resin and subjected to the staining procedure to examine this possibility. A comparison of resin distributions before and after the staining procedure indicated no substantial differences due to the staining treatment (Fig. 6). Furthermore, vigorous agitation of the fiber suspended in the staining solution does not cause a strong increase in big or small resin objects, and the variation between the distributions is at a level that was observed for identical samples.

The adapted staining procedure, which had shown to be suitable for staining both wet and dry (post-dryer) resinated fiber from the blowline, was then used to survey differing MDF production parameters at several local mill facilities. This had a two-fold objective, to apply the fiber staining procedure (adapted method) across a range of mills, but also to observe what effect relatively simple changes in MDF processing conditions have on resin distribution in panels. Individual mills were instructed to produce two sets of panels, suggested to be relatively "good" and "poor" panels, by varying processing conditions, which likely affect resin distribution in MDF panels. Here, suggested parameters to vary,

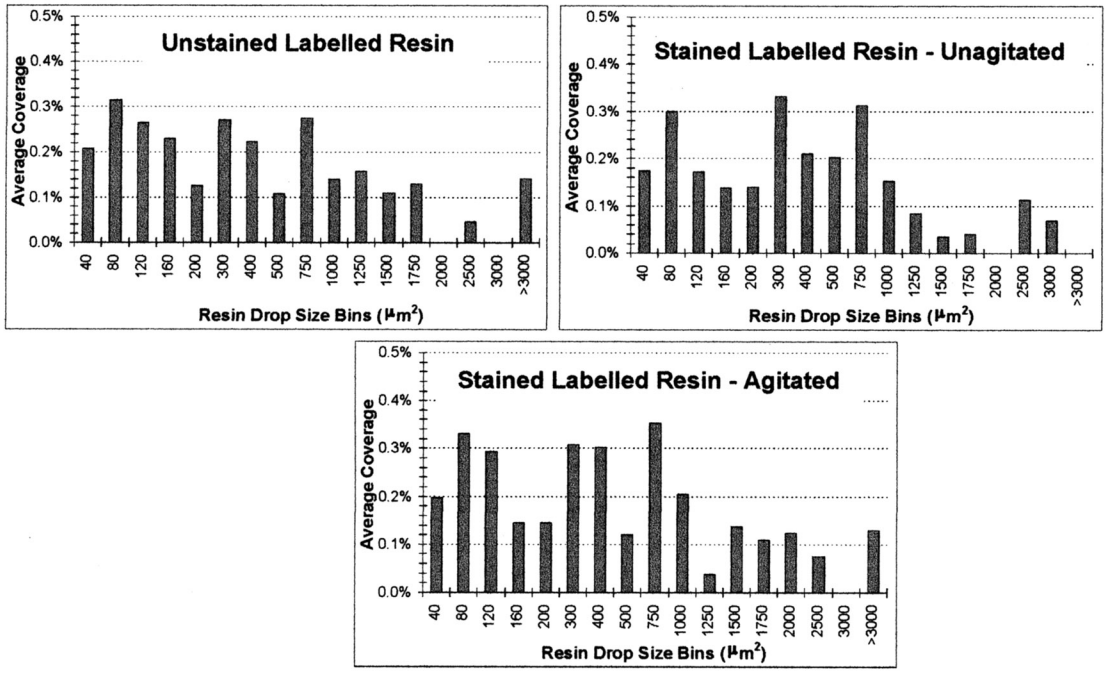

FIG. 6. Resin distribution results comparing unstained fiber (with labelled resin) with stained fiber, either unagitated or agitated during the staining process.

which have been reported by others as well as from our own research, included resin injection nozzle type and position (Loxton et al. 2003); pressure drop across nozzle (Robson 1991), and mass flow parameters such as amount of water (Kamke et al. 2002), while all other production variables were maintained constant.

A range of resinated fiber was analyzed, varying in resin type and content, sampling position, and change in process variable (Table 1). Confocal microscopy analysis of stained fiber sets from each mill revealed variations in resin coverage of fiber (Fig. 7). Overall, the results indicated a variation in average resin coverage, ranging between 3 and 16%. These coverage values are not absolute values, but were dependent on the corresponding stained unresinated fiber, which was used as a reference for thresholding. Individually, for each mill, there were differences apparent between samples. The highest observed difference (10%) was seen for Mill 2, in which the lower resin loading gave less resin coverage. For Mill 3 a similar decrease in coverage from *ca.* 11% to 3%, was observed over the three samples examined. This difference was due to a consecutive manipulation of blowline conditions at a constant resin loading (8%). For the other mills, relative differences of *ca.* 4% were observed between their respective samples with the differences attributable to variations around the resin nozzle. The observed changes in coverage observed for each mill were in line with expectations based on previous work (Kamke et al. 2002; Loxton et al. 2003).

Also shown in Fig. 7 is the result for fiber that was stained two weeks after being sampled at Mill 1. Freshly stained fiber (1–2) had a comparable coverage (6.2%) to that of the same fiber sample stained two weeks later in the laboratory (6.6%). This result indicates it may not be necessary to immediately stain freshly resinated fiber using this staining procedure. In the case of Mill 4, this finding presented an opportunity for resinated fiber to be stained solely after reaching the authors a week later, rather than necessitating immediate staining of freshly resinated fiber at the mill.

The investigation of resin drop size distributions also revealed considerable variations between samples of certain mills (Figs. 8 and 9). Evident for Mill 2, were droplet-size distributions that were similar between the two samples, except for the proportion of "very large" droplets (Fig. 8). The sample with the greater proportion of very large droplets had a higher resin loading, which appears to influence only large resin features. Mill 3 fiber samples had shown a variation in coverage values. In this case, changes to the blowline conditions had been made. Similarly, differences were also evident in resin distribu-

TABLE 1. *Resin coverage values for stained fiber from MDF Mills.*

Mill	Processing Parameter Change*	Fiber Set	Resin Loading (%)	Fiber Sampled	Resin Coverage (%)
1	nozzle	1 (a)	10	Cyclone	7.4
	nozzle	1 (b)	10	Pendistor	5.2
	nozzle	2 (a)	10	Cyclone	6.2
	nozzle	2 (a)	10	Cyclone[a]	6.6
	nozzle	2 (b)	10	Pendistor	10.4
2	resin loading	1	12	Cyclone	15.5
	resin loading	2	7.7	Cyclone	5.3
3	blowline	1	8.5	Cyclone	11
	blowline	2	8.5	Cyclone	6.3
	blowline	3	8.5	Cyclone	3.0
4	nozzle	1	10	Cyclone[b]	10.0
	nozzle	2	10	Cyclone[b]	6.0

* Limited information is presented here.
a = stained 2 weeks after sampling.
b = stained 1 week after sampling.

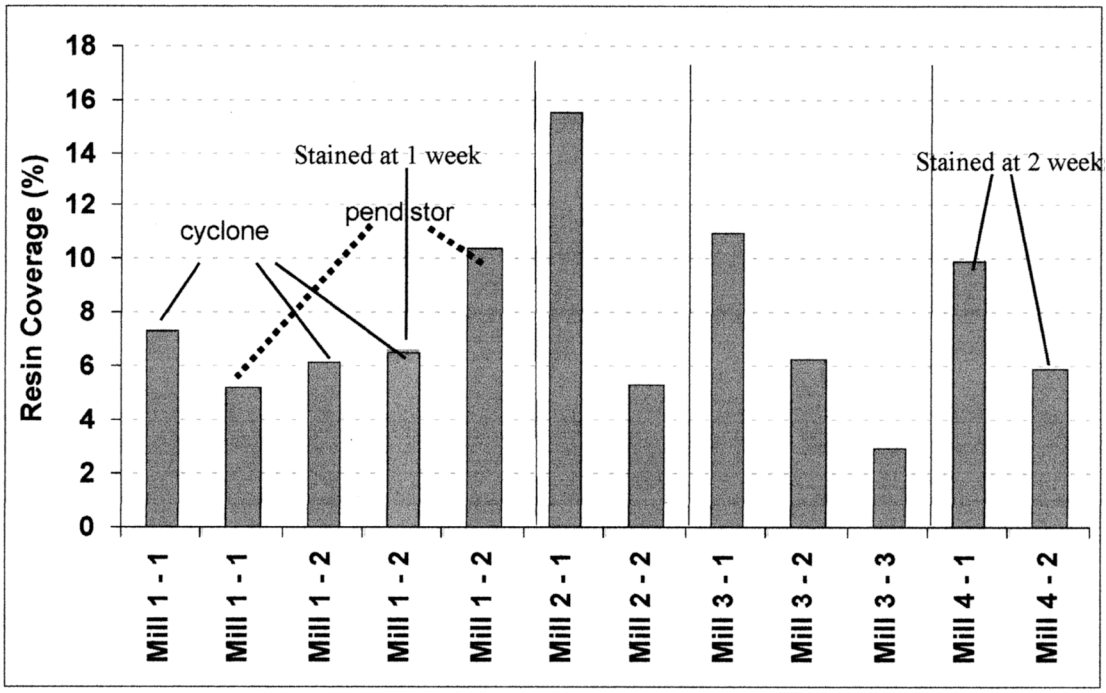

FIG. 7. Comparison of resin coverage values for resinated fiber samples obtained from various mills, trials, and sampling locations (refer to Table 1).

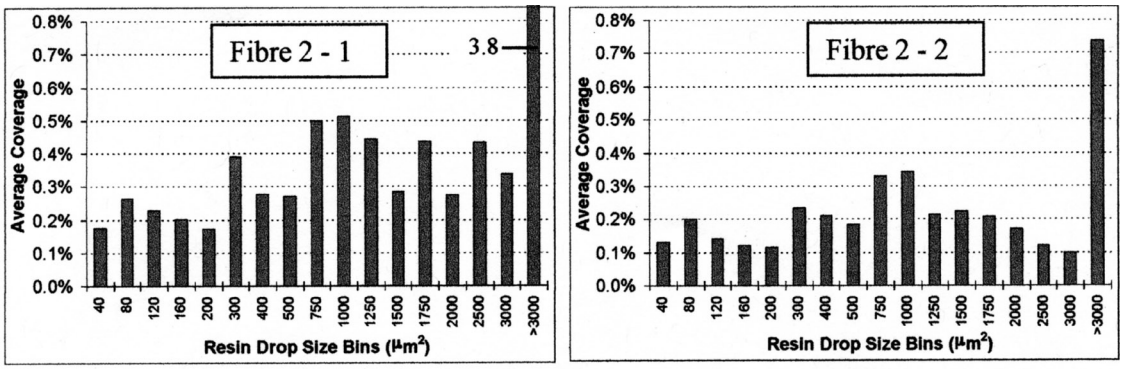

FIG. 8. Resin droplet distributions on fiber sampled at Mill 2.

tions between samples (Fig. 9). The sample having higher coverage was associated with greater proportions of larger drops compared to the other samples having lower coverage.

The results from the four mills show that the adapted staining procedure was able to distinguish differences in stained resinated fiber from a variety of fiber and resinating conditions.

The staining method was able to distinguish between changes in resin loading, but was also able to determine differences in resinating conditions based around relatively simple changes in blowline and resin injection nozzle parameters. In the case of Mill 3, which used a relatively lower resin loading (*ca.* 8%), discrete differences in resin coverage and distribution

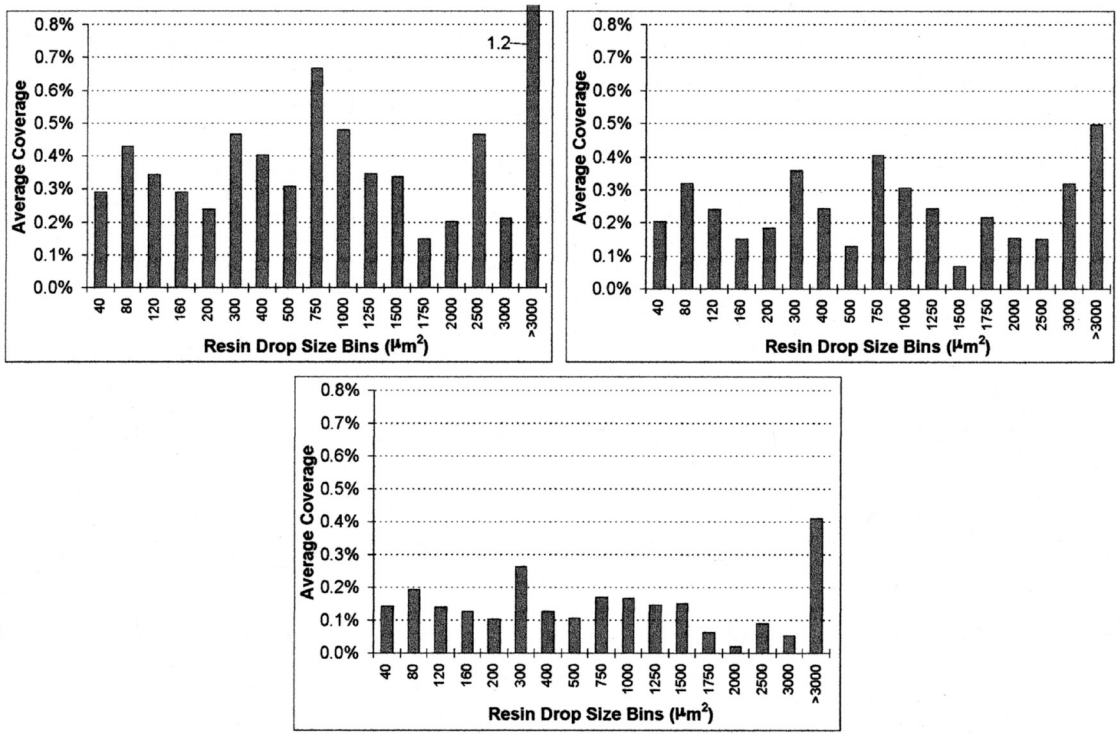

FIG. 9. Resin droplet distributions on fiber sampled at Mill 3.

were obtained with changes made to blowline parameters. Similarly, a distinction in coverage values due to nozzle set-up was also achieved for Mill 4, which used a higher resin loading (10%). This result should be compared with that obtained for dry-blending at higher resin loading (Fig. 4). It is also worth noting the scale of these relative differences for resin coverage and distribution were greater for changes in resin loading than differences in blowline or nozzle parameters.

It is expected this technique is similarly applicable with other amino-based resins such as melamine containing MUF resin, and readily applies to other types of panel furnish such as wood particles, strands, or flakes. Furthermore, the possibility of staining fiber sometime after sampling from the mill suggests that the fiber staining treatment does not need to be done immediately, but can be left for a week or more. Therefore, routine fiber sampling during panel manufacture could aid identification of the cause of panels failing quality control.

The changes that had been made to the original method had improved the staining process and consequently increased contrast between stained resin and fiber when CLSM was used. An attempt was made to apply the modified staining method to conventional fluorescence microscopy, for which the original method had been devised. Dry-blended fiber, stained using the adapted procedure, was analyzed using fluorescence microscopy. Shown in Fig. 10 are images of both unresinated and resinated fiber, in which resin features appear reddish in color compared to the yellow-green of fiber. The resin features have, however, very little contrast against the fiber background that made image analysis difficult, and inconclusive resin coverage and distribution results were achieved. Ap-

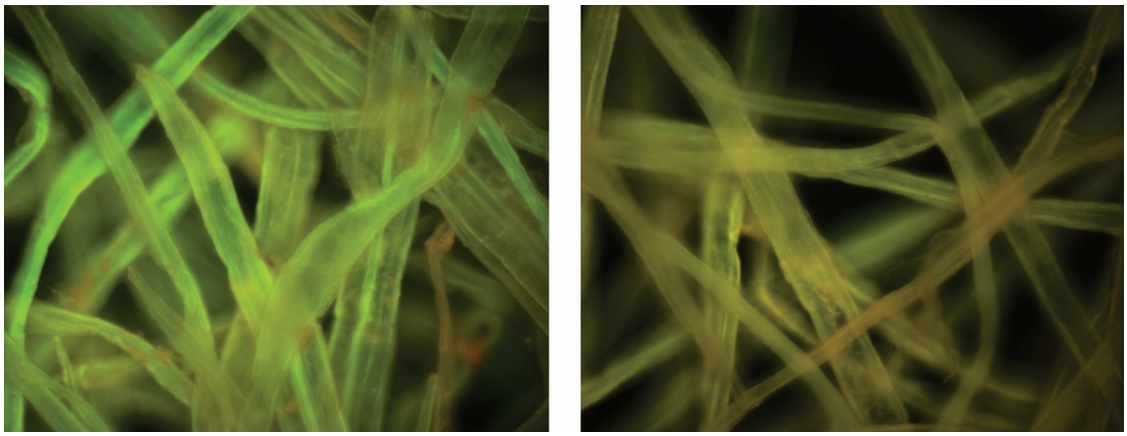

FIG. 10. Fluorescence microscope images of resinated and unresinated fiber stained with the adapted method.

parently the achieved improvements to the staining procedure are specific to CLSM and can not be transferred to conventional microscopy.

CONCLUSIONS

A staining method coupled with confocal microscopy was developed to determine amino-based resin coverage and distribution on dry-blended and blowline-resinated MDF fiber. The procedure does not require any pretreatment of resin, and resinated fiber can be sampled from any point during the MDF process during normal operation. Fiber samples can be stained either fresh or up to two weeks after manufacture. For laboratory dry-blended fiber resinated using differing spray atomization regimes, resin coverage and distribution results determined on stained fiber were found to be comparable to those from fluorescently labelled UF resin.

Stained blowline-resinated fiber was analyzed to compare resin distributions created at several MDF mills during typical running conditions. In each case, differences in resin coverage and distribution were observed for changes to resin loading and to blowline and nozzle parameters. The potential of this method has been demonstrated in that the staining procedure could be used as a tool to assess the influence of varying process conditions for line improvements, quality control, and trouble-shooting applications.

ACKNOWLEDGMENTS

The authors would like to thank the New Zealand Composite Panels Research Group for supporting this work. We would also like to thank staff at participating mills for their assistance in providing stained fiber.

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