PRESSURE TREATMENT OF SOUTHERN PINE POLES WITH CHLOROTHALONIL IN HYDROCARBON SOLVENT[1]

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ABSTRACT

A series of end-sealed southern pine pole stubs was treated with chlorothalonil in hydrocarbon solvent. Over sixty sections were treated using various combinations of initial seasoning (kiln-drying, steam-conditioning), treating temperature (ambient, heated), initial air pressure, and final conditioning (steam flash + vacuum, expansion bath + vacuum, steam distillation + vacuum). Disks removed from the treated sections were analyzed for preservative gradient. Results are discussed in terms of treatability and the impact of treating variables on preservative retention, penetration, and gradient shape. These data indicate that acceptable treatment of southern pine pole stock can be obtained with either steam-conditioned or kiln-dried pole stock. The use of heated solution appears to be preferred over ambient solution, although acceptable treatment can be achieved at ambient conditions.

Keywords: Chlorothalonil, pressure treatment, seasoning, conditioning, retention, penetration, preservative gradient.

INTRODUCTION

Chlorothalonil [CTL] (2,4,5,6-tetrachloro-1,3-benzenedicarbonitrile) has been used widely as an agricultural fungicide in the United States and is registered for use on over 35 different crops. CTL also finds use as a mildewcide in paints and as a mold and sapstain preventive for sawn lumber (Laks et al. 1991; Laks et al. 1992b; Woods and Klaver 1992). Its broad spectrum efficacy, minimal environmental impact, and low mammalian toxicity have promoted research on its use as a ground contact wood preservative (Laks and Woods 1992; Woods and Bell 1990). CTL was moved to standard by the AWPA in 1993 under the P8 Standard for Oil-borne Preservatives (AWPA 1993). Several studies have dealt with the efficacy of chlorothalonil, alone or in combination with other biocides, for protecting wood against attack by termites (Creffield and Chew 1993a, 1993b; Grace et al. 1992, 1993; Woods 1992a, b; Woods et al. 1994a), and marine borers and fungal colonization by decay organisms (Cookson 1994; Laks et al. 1992a). Typical ground contact stake test results have been published (Barnes 1993), and CTL has been widely recommended as a potential pole/crossarm preservative (Laks and Woods 1992; Preston 1986, 1992; Preston et al. 1985, 1987; Woods et al. 1994b). Within AWPA, commodity standards require that treatability with a preservative be demonstrated before the candidate preservative system can be listed in the requisite commodity standard(s).

This study was designed to investigate basic treating process parameters for CTL in P9 type A (AWPA 1993) hydrocarbon solvent. The objective of Phase I was to determine the effect of seasoning method (kiln-drying, steam-conditioning) and treating parameters (treatment

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temperature, initial air pressure) on preservative gradients for southern pine pole-sized material. Phase II examined the effect of post-treatment conditioning on the resultant preservative retention and penetration. The final phase (Phase III) verified the previous results in full-scale pilot plant charges.

TABLE 1. *Moisture content of samples after seasoning.*

Phase: conditioning method	Avg moisture content ± std deviation (%)
Phase I: Kiln-dried	18.7 ± 1.8
Phase I: Steam-conditioned	45.8 ± 6.4
Phase II: Kiln-dried	16.3 ± 3.5
Phase II: Steam-conditioned	66.4 ± 9.9

METHODS AND MATERIALS

Initial conditioning of pole stock

Green southern pine (*Pinus* spp.) roundstock with a nominal diameter of 190 mm was procured from a local pole yard in 2.4-m lengths. These pole stubs were end-sealed and subsequently seasoned by kiln-drying or steam-conditioning prior to treatment. Kiln-drying was accomplished in a steam-heated batch kiln at a constant wet bulb temperature of 52°C and a maximum dry bulb temperature of 77°C. Steam-conditioning was achieved by closed steaming the stubs in a treating retort at 118°C for 9 h followed by the application of vacuum at ≥ -94 kPa for 1 h. In Phase I and II, each stub was sectioned into 460-mm-long specimens for treatment plus an adjacent disk used for moisture content determination by oven-drying (od). In Phase III, samples were trimmed to 2.2 m and treated full length. Each specimen was weighed, measured, and end-sealed with phenol resorcinol resin prior to treatment. Moisture contents at the time of treatment are given in Table 1 for the material treated in Phases I and II.

Phase I treatments

Phase I samples were treated by an empty-cell process in an invertible vertical treating cylinder. The retort was divided into halves by a perforated plate welded at the midpoint of the cylinder. The lower portion contained steam coils for heating the treating solution. In a typical treatment charge, the conditioned sample was placed in a sample holder consisting of two perforated plates attached by carriage bolts. Rubber disks were inserted between the sealed ends of the sample and the end plates, and the plates were drawn together until the assembly was snug. The sample was placed in the upper half of the treating cylinder, and the bottom half was charged with treating solution. An initial air pressure of 0, 69, 138, 276, or 414 kPa (gauge) was applied and held for 30 min, and the cylinder was inverted, thus submerging the sample. Air pressure was applied through a weighed scale tank containing preservative solution such that the maximum pressure of 1,034 kPa (gauge) was reached in 30 min. Pressure was maintained until the desired gross injection or the refusal point[2] was reached. The cylinder was then flipped over, vented to atmospheric pressure, and the preservative solution removed. A one-hour final vacuum at maximum intensity completed the treatment. Samples were treated with the solution at either ambient temperature or at 62-86°C (average = 77°C at maximum pressure). Solution retention was calculated from the weight gain data. A nominal 4% solution of chlorothalonil dissolved in AWPA P9 type A hydrocarbon solvent (AWPA 1993) was used for all treatments. Characteristics of the solvent are shown in Table 2.

Phase II treatments

The Phase II study investigated the effects of post-treatment conditioning. Specimens were treated as above using a 276 kPa (gauge)

[2] The refusal point in this study was taken as that point where no additional gross solution absorption was recorded in three successive measurements at five-minute intervals. In the Phase I study, one charge of kiln-dried stock at ambient treatment was treated to refusal and the duplicate charge was treated to 224 kg/m^3 gross solution injection or refusal, whichever came first. The remaining duplicate charges in Phases I & II were all treated to refusal.

TABLE 2. *Characteristics of the whole oil carrier (AWPA P9 type A).*

Test (ASTM standard)	Results
Color (D1500)	0.5
Density at 15°C (D1298/D4052)	959.3 kg/m^3
Flash point, PMMC (D93)	101.7°C
Viscosity at 40°C (D445)	3.65×10^{-6} m^2/s
Distillation (D86)	°C
Initial boiling point	204.4
48% recovery	254.4
49% recovery	260.0
90% recovery	383.3

initial air pressure. After treating, samples were conditioned by either steam + vacuum (SV), expansion bath (EB), or steam distillation (SD). After venting to atmospheric pressure and removing the preservative from the retort, the SV cycle consisted of flooding the steam coils with water, raising the temperature to 118°C in 30 min, and closed steaming the treated samples at 118°C for an additional 30 min. The cycle was completed by the removal of the water and the application of a final vacuum at maximum intensity for 60 min. In the EB cycle, preservative solution was retained in the retort after venting to atmospheric pressure. The solution was heated to 93°C, and the sample was allowed to condition in the hot fluid for 1 h. The SD cycle was similar to the SV cycle except that the steaming water was maintained in the retort during the application of a vacuum for 90 min. During the vacuum period, the steam to the coils was shut off and reapplied only if the temperature fell below 77°C during the first hour of vacuum.

Phase III treatments

Schedules were verified in Phase III of the study. In this phase, kiln-dried and steam-conditioned pole stubs nominally 2.2 m long were treated in a pilot plant using heated solution and 0 kPa (Lowry) or 276 kPa (Rueping) initial air pressure. Final conditioning was accomplished using the steam distillation cycle described above. Except for the first charge, all treatments were discontinued after a 90-min pressure period.

TABLE 3. *Average net solution injection results obtained in Phase I with kiln-dried and steam-conditioned stock.*

Initial air pressure (kPa, gauge)	Net retention (kg/m^3)	Refusal time (min)	Net retention (kg/m^3)	Refusal time (min)
	Ambient		Heated	
Kiln-dried stock				
0	401.3[1] (198.6)	85 (55)	376.1	125
69	285.3[1] (201.1)	115 (75)	306.7	130
138	251.0[1] (189.8)	125 (85)	275.7	128
276	166.6[1] (151.4)	150 (105)	196.2	135
414	134.4	110	103.0	80
Steam-conditioned stock				
0	261.3	73	259.1	25
69	204.3	70	251.8	53
138	203.3	63	263.2	38
276	140.5	63	174.0	43
414	96.1	58	96.4	43

[1] Represents a single charge treated to refusal or a charge treated to 224 kg/m^3 (in parentheses) gross injection. The remaining data in the table represent the average of duplicate charges treated to refusal.

Determination of preservative gradients

Following treatment, a central disk was taken from each treated sample. A pie-shaped wedge was sawn from each disk, segmented radially into assay zones (outer 12.5 mm, next 37.5 mm [AWPA assay zone], next 25.4 mm, next 25.4 mm), ground, and analyzed using X-ray fluorescence spectroscopy. Assay values were normalized to a 5% treating solution concentration by multiplying the assayed retention value by the ratio of a 5% to actual treating solution concentration. For comparison purposes, the slopes of the gradients were calculated using a linear regression fit.

RESULTS AND DISCUSSION

Phase I
Effect of treating parameters

Average net solution injection data from the Phase I study are shown in Table 3 for the kiln-dried and steam-conditioned stock. Typical gross solution injection curves are shown in Fig. 1.

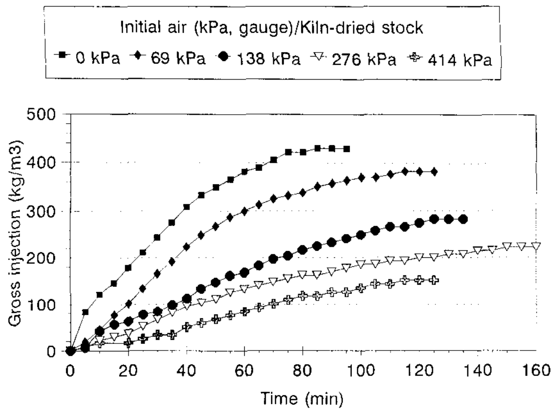

FIG. 1. Typical gross solution injection curves at different initial air pressures for kiln-dried stock treated to refusal at ambient temperature.

As expected, net solution absorption decreased with increasing initial air pressure. As shown in Fig. 2, steam-conditioned samples treated to a lower net solution retention than did kiln-dried sections. The effect of heating on solution injection was negligible for kiln-dried stock resulting in an overall increase across all initial air pressures of only 1.2%. Heating preservative solution had a slightly greater effect on the net absorption of steam-conditioned stock with a resultant increase in absorption of 16.1% across all initial air pressures.

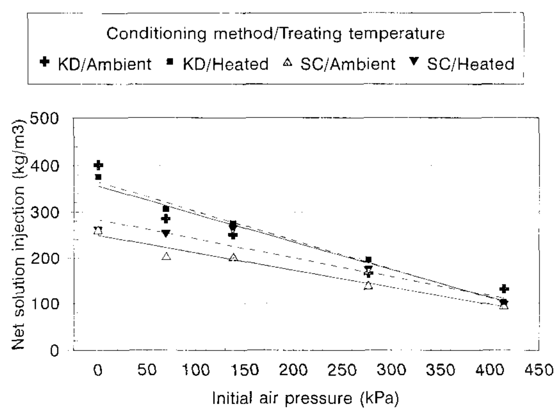

FIG. 2. Effect of initial air pressure and temperature on net solution injection of chlorothalonil in hydrocarbon solvent in kiln-dried (KD) and steam-conditioned (SC) southern pine pole stock treated to refusal [dashed lines = heated].

TABLE 4. *Effect of initial conditioning method, treating temperature, and initial air pressure on chlorothalonil gradients in southern pine (Phase I).*

Conditioning method	Treatment temperature	Initial air pressure (kPa)	Gradient slope ([kg/m³]/mm)	Average
Kiln-dried	Ambient	0	−0.007	
		69	−0.024	
		138	−0.039	−0.025
		276	−0.037	
		414	−0.020	
	Heated	0	−0.093	
		69	−0.066	
		138	−0.025	−0.050
		276	−0.046	
		414	−0.021	
Steam-conditioned	Ambient	0	−0.057	
		69	−0.032	
		138	−0.054	−0.038
		276	−0.037	
		414	−0.009	
	Heated	0	−0.090	
		69	−0.034	
		138	−0.009	−0.039
		276	−0.033	
		414	−0.027	

The effect of the Phase I processing parameters on preservative gradients is shown in Figs. 3 and 4 and Table 4. With respect to initial air pressure, the gradients tended to flatten out as the initial air pressure increased. This indicates a more uniform treatment across the pole cross section. With kiln-dried stock across all initial air pressures, the gradients obtained with heated preservative were generally steeper and higher in magnitude than those obtained with unheated treating solution (Fig. 4a). With steam-conditioned stock, little difference in slope or magnitude was noted between treating temperatures when gradient averages across all initial air pressures were compared (Fig. 4a). Gradients for the two initial seasoning methods were essentially equivalent when averaged across all initial air pressures (Fig. 4b). The average steam-conditioned gradient was slightly lower and steeper than that found for kiln-dried stock (Fig. 4b). When averaged across treating temperature, heated solution

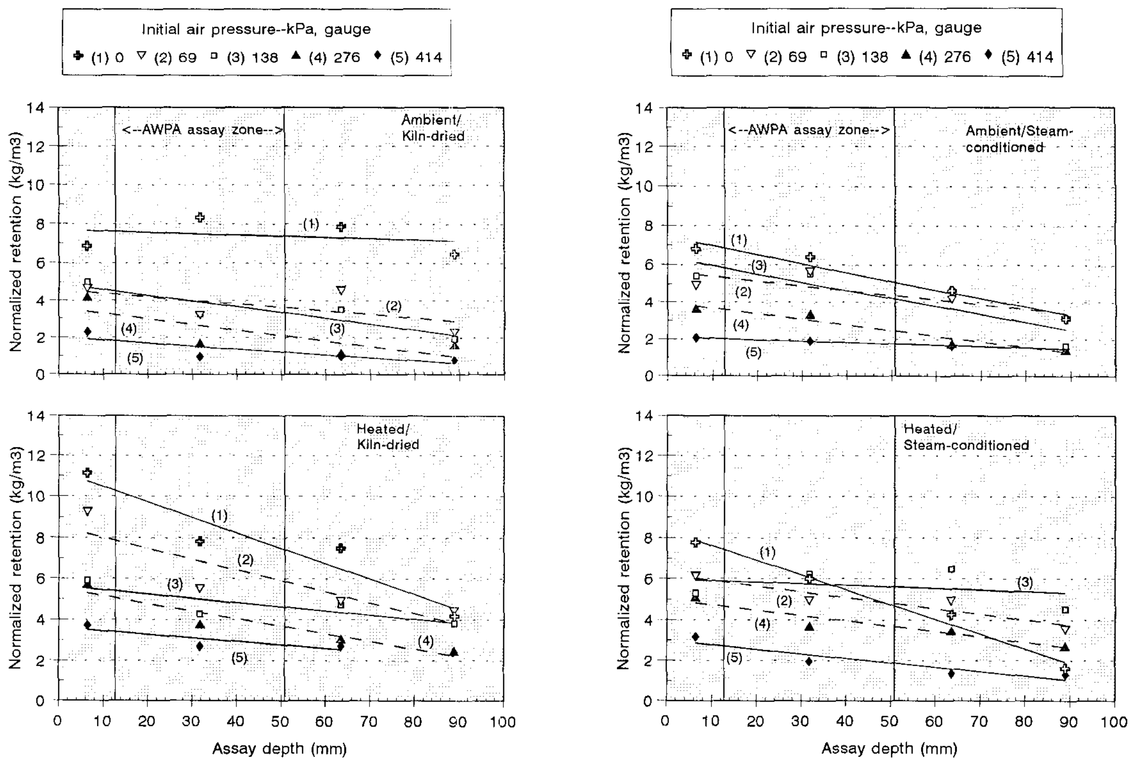

FIG. 3. Chlorothalonil gradients as a function of initial conditioning method, initial air pressure, and treating temperature for southern pine pole sections treated to refusal. [Dashed lines = heated; solid lines = ambient].

yielded gradients of greater magnitude, but the slopes at both temperatures were essentially equivalent (Fig. 4b).

A comparison of chlorothalonil gradients obtained in this study are compared with gradients for other P9 type A oilborne preservative systems found in the literature in Fig. 4c. The chlorothalonil gradients obtained are generally flatter than those for pentachlorophenol (Kelso and Parikh 1976; Rogers and Kelso 1958) and are of the same shape as those found with copper naphthenate (Barnes and Hein 1988). These comparisons suggest that treatment with chlorothalonil in P9 type A solvent should result in gradients appropriate for commercial use. These data would indicate that either method of initial seasoning will yield acceptable treatment of southern pine pole stock. The use of heated solution appears to be preferred over ambient solution, although acceptable treatment can be achieved at ambient conditions.

Phase II
Effect of post-treatment conditioning

The methods for final conditioning of the stock in this preliminary study varied slightly from what might be done in commercial practice. In this phase of the study, no final vacuum was applied between the maximum pressure period and the final conditioning. In commercial practice, it is commonplace to apply this vacuum to reduce drippage. Because of environmental/pollution considerations, the volume of oil in the bottom of the treating retort arising from this vacuum is then pumped out of the retort so that it will not be mixed with any steaming condensate that might be generated. This step facilitates the cleanup of

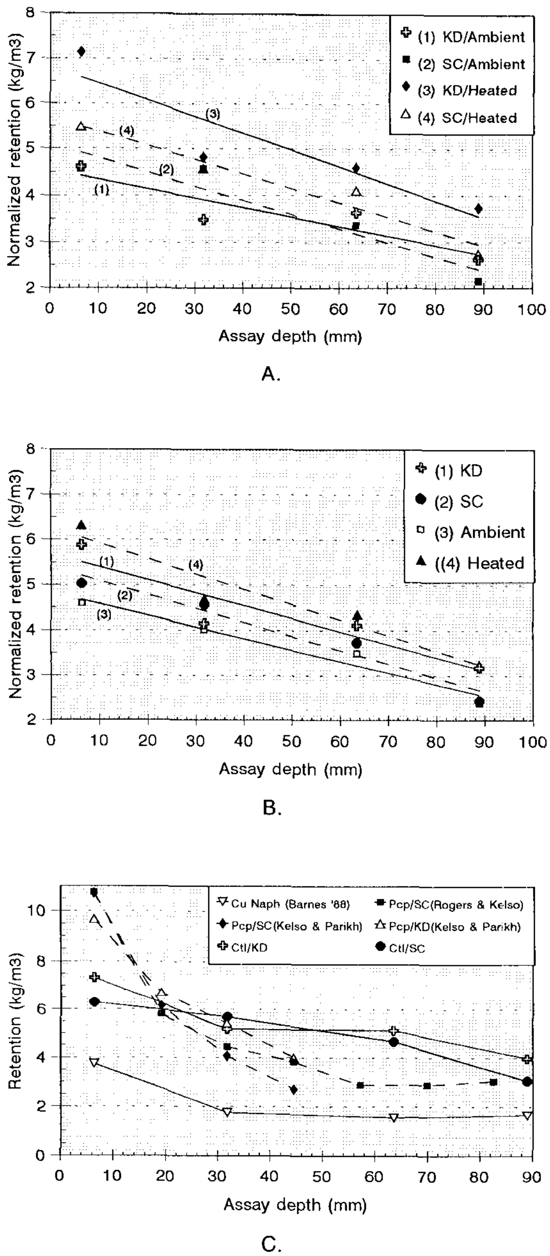

FIG. 4. A comparison of preservative gradients for: (A) conditioning method and treating temperature combinations averaged across all initial air pressures; (B) for conditioning method averaged across both treating temperatures or for treating temperature averaged across both conditioning methods; and (C) a comparison of chlorothalonil (Ctl) gradients with published literature values for pentachlorophenol (Pcp) and copper naphthenate (Cu-Naph) in P9 type A solvent.

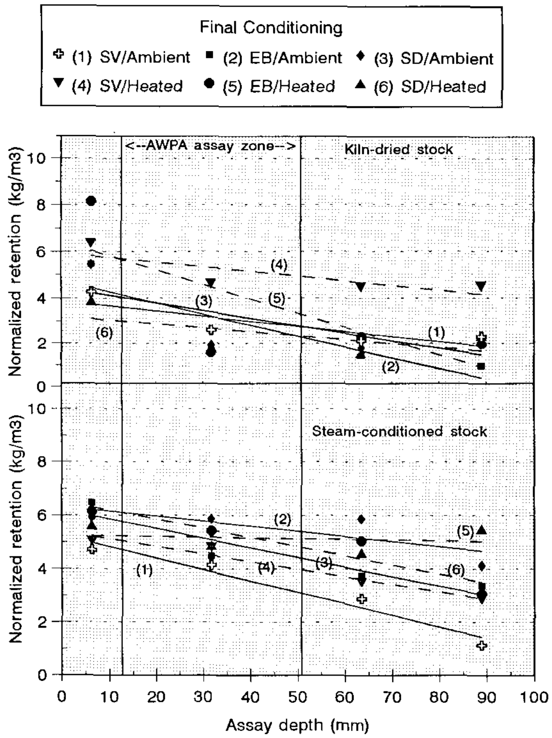

FIG. 5. A comparison of gradients obtained in the post-treatment conditioning study (Phase II) for steam + vacuum (SV), expansion bath (EB), and steam distillation (SD) methodologies.

any process water. It should also be noted that an expansion bath is not generally used for the final conditioning of southern pine. In commercial practice, a vacuum is often applied during the heating phase. This would mean that the expansion bath is essentially a final Boulton cycle. Boultonizing is normally performed only on thin sapwood species because the volume of water generated in thick sapwood species like the pines would exceed the condensing capacity of most Boultonizing operations.

Gradients and gradient slopes from the Phase II investigation of final conditioning methods are shown in Fig. 5 and Table 5. When these gradients are analyzed from the AWPA assay zone inward, the following trends are evident. Gradients for steam-conditioned stock tended to be higher than those for kiln-dried stock,

TABLE 5. *Effect of initial and final conditioning methods on chlorothalonil treatment and gradients in southern pine (Phase II).*

Conditioning			MC at treat- ment	Time to	Net retention (kg/m^3)	Gradient slope ($[kg/m^3]/mm$)	
Initial	Final	Treating temperature	(%)	refusal	(min)	Individual	Avg
Kiln-dried	Steam + vacuum	Ambient	20.4	125	85.7	−0.029	−0.027
		Heated	20.9	88	108.4	−0.025	
	Expansion bath	Ambient	15.0	123	108.7	−0.061	−0.070
		Heated	14.3	80	126.2	−0.078	
	Steam distillation	Ambient	13.0	105	86.7	−0.042	−0.032
		Heated	12.8	48	81.2	−0.022	
Steam-conditioned	Steam + vacuum	Ambient	56.6	55	93.3	−0.054	−0.045
		Heated	54.7	38	92.6	−0.035	
	Expansion bath	Ambient	51.8	53	125.1	−0.045	−0.044
		Heated	54.6	33	121.3	−0.043	
	Steam distillation	Ambient	50.2	55	111.9	−0.024	−0.014
		Heated	30.0	33	92.0	−0.003	

but the slopes were identical. With respect to temperature, gradient slopes were equivalent, with gradients for stock treated at 77°C slightly above those for treatment at ambient conditions. Material that had been conditioned by steam distillation yielded the highest and flattest gradients, while that conditioned by expansion bath had both the lowest and steepest gradients. In this particular study, the application of steam in the final conditioning process would seem to be preferred.

Phase III
Schedule verification study

Poles from this phase were extremely clean, light-colored, and drip-free when removed from the cylinder. Similar physical characteristics have been shown for pentachlorophenol-treated southern pine poles conditioned using steam distillation (Kelso and Parikh 1976; Thompson 1974). Gradients and retentions are shown in Fig. 6 and Table 6. Lowry treatment yielded nearly linear gradients that were slightly steeper compared to those for Rueping-treated material. For a target of 4.8 kg/m^3 in the AWPA assay zone, an initial air pressure less than 276 kPa should be used. At equivalent initial air pressures, kiln-dried stock yielded better results than steam-conditioned stock.

SUMMARY AND CONCLUSIONS

Based on this preliminary study, no problems with treating southern pine pole stock with chlorothalonil in hydrocarbon solvent were encountered. The use of chlorothalonil in hydrocarbon solvent is amenable with traditional initial seasoning methods, empty-cell treating schedules, and post-treatment conditioning processes. Best results were obtained when treating steam-conditioned stock with heated preservative using an initial air pres-

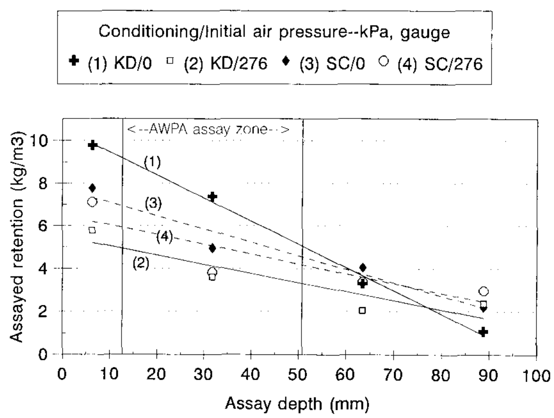

FIG. 6. Preservative gradients obtained for kiln-dried (KD) and steam-conditioned (SC) stock treated with heated preservative at two initial air pressures in the schedule verification study (Phase III).