Reformulation of Alkaline Copper Quat for Enhanced Copper Leaching Resistance

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To ensure the environmentally safe usage of copper amine-based wood preservatives in aquatic environments, it is necessary to minimize copper leaching from treated wood. In this study, alkaline copper quat (ACQ) was reformulated to enhance resistance to copper leaching by adjusting the proportions of copper, didecyldimethylammonium chloride (DDAC), and mono-ethanolamine (Mea) solvent. The copper proportion in the formulation was decreased 40% while maintaining the total retention of active ingredients through increasing DDAC. The molar ratio of Cu to Mea in the formulation was then adjusted from 1:4 to 1:2.75. This reformulation shortened the time to copper stabilization from 15 to 6 days, and reduced cumulative copper leaching by 75%, compared to a control formulation. These fixation properties were further improved with just a 30-min hot-air post-treatment at 100 °C. Wood treated with the reformulated ACQ exhibited comparable performance in biological efficacy against fungi and termites compared to wood treated with commercial ACQ.

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INTRODUCTION

Copper amine-based preservatives, including alkaline copper quat (ACQ), copper azole (CUAZ), and bis-(N-cyclohexyldiazeniumdioxy)-copper (CuHDO), have become prevalent in Korea following the ban on the use of chromated copper arsenate (CCA)-treated wood. These preservatives offer biological efficacy against fungi and termites comparable to CCA, with relatively fewer health concerns (Evans 2003; Westin *et al.* 2010; Temiz *et al.* 2014). However, they face challenges with poor copper fixation in wood, leading to significant copper leaching (Tascioglu *et al.* 2005, 2008; Ung and Cooper 2005). This leaching remains a concern, particularly in aquatic environments, as copper is highly toxic to both freshwater and marine biota even at low concentrations (Eisler 1998). Considering the excessive copper leaching and its high aquatic toxicity, the aim of this research field was to minimize copper leaching from wood treated with such preservatives for environmentally safe use in aquatic settings.

Poor copper fixation from wood treated with copper amine-based preservatives can be attributed primarily to the high copper content compared to CCA. In copper-mono-ethanolamine (Cu-Mea)-based preservatives, copper impregnated within wood cells are present as various Cu-Mea complexes. The dominant $[Cu(Mea)_{2-H}]^+$ species binds to the carboxylic groups of hemicellulose and phenolic hydroxyl groups within wood cells, and releases one Mea molecule (Zhang and Kamdem 2000; Lee and Cooper 2010a,b). Unbound copper within cells can precipitate as basic copper carbonates when the pH within impregnated wood decreases (Jiang and Ruddick 2004; Lee and Cooper 2012). The number of acidic groups within the treated wood that can chemically adsorb copper is significantly lower compared to the number of copper-amine complexes. In addition, not all the remaining copper, which is not chemically adsorbed, converts into insoluble copper precipitates, as the pH of the treated wood slowly decreases after treatment (Lee and Cooper 2012). These factors could lead to inevitable copper leaching from the treated wood.

Some studies have examined the effects of preservative proportion adjustment on copper leaching primarily focusing on a formulation of ACQ (composed of copper, mono-ethanolamine as a solvent, didecyldimethylammonium chloride (DDAC) as a co-biocide). At similar retentions of active ingredients (CuO + DDAC), the preservative with lower copper proportions resulted in an increase in fixation and a decrease in leaching (Pankras *et al.* 2009). A previous study showed that in Cu-Mea solution, significant copper was precipitated when the solution approached neutral pH at moderate ratios of Cu to Mea (Lee and Cooper 2010b). However, at a high ratio of Cu to Mea (*e.g.*, 1:70), *in vitro* Cu precipitation was not observed, which is most likely due to inhibition of Cu precipitation when free amine prevails over hydroxyl ions. These results implied that the reformulation of the Cu to Mea ratio within preservatives might promote the precipitation within wood and reduce copper leaching. Other studies have investigated post-treatment methods to minimize copper leaching from treated wood. These treatments primarily involve the application of high temperatures, which facilitate the conversion of soluble copper species into insoluble forms (Ung and Cooper 2005; Tascioglu *et al.* 2008; Yu *et al.* 2009, 2010; Ye and Morrell 2015). However, there has been insufficient research evaluating the combined effects of individual reformulation factors on minimizing copper leaching. Furthermore, the potential impact of these reformulations on biological efficacy remains unknown.

This study was carried out to enhance the copper leaching resistance of wood treated with copper amine-based preservatives through a series of reformulations. ACQ was chosen as the preservative for this study due to its dominance in the wood preservative market in Korea and its well-characterized formulation. Laboratory tests on copper fixation and leaching were conducted to evaluate the effects of the reformulation. To further minimize copper leaching, the optimal post-treatment parameters for wood treated with reformulated ACQ were identified. Additionally, the biological efficacy of wood treated with reformulated preservatives was evaluated with laboratory-scale tests. This study highlights the evaluation of combined effects of individual factors on minimizing copper leaching and their potential impact on biological efficacy.

EXPERIMENTAL

ACQ Reformulation and Preparation

The relative proportions of Cu, DDAC, and Mea in a formulation of control ACQ were set with the following conditions: a weight ratio of Cu to DDAC of 1.6:1 and a molar ratio of Cu to Mea of 1:4, which is consistent with the formulation of commercial ACQ in

Korea. The preparation of control ACQ followed the slightly modified method described in previous research (Pankras *et al.* 2009). Briefly, a Cu-Mea stock solution (containing 8% elemental copper by weight) and a 5 % DDAC solution (w/w) were separately prepared and then mixed. The Cu-Mea stock solution was prepared by dissolving basic copper carbonate ($CuCO_3 \cdot Cu(OH)_2$) in a pre-warmed Mea solution, while the DDAC solution was prepared by diluting pure DDAC.

The reformulation of ACQ was first conducted by modulating the relative proportion of Cu to DDAC. The copper content was decreased 10 to 40% by weight in the Cu-Mea stock solution while keeping the molar ratio of Cu to Mea at 1:4. To maintain the total retention of active ingredients, the DDAC content was increased to compensate for the copper retention reduction. Further reformulation of the copper-reduced preservative was performed by adjusting the molar ratio of Cu to Mea from 1:4 to 1:2.75.

Sample Preparation

Treated samples for fixation and leaching tests were prepared as previously described (Tascioglu *et al.* 2005). In brief, 19 mm × 19 mm × 19 mm cubes were cut from air-dried Radiata pine (*Pinus radiata*) sapwood. The blocks were treated using control ACQ and reformulated preservatives by the full-cell process. The pressure treatment involved placing the wood specimens in a small pressure vessel, followed by a 15-min pre-evacuation at a vacuum pressure of 760 mmHg, and then applying a pressure of 14 kg/cm² until the refusal point. The treated specimens were wrapped with disposable wrap and plastic foil and incubated at 21 °C. Samples for fixation and leaching tests were randomly taken at different times. For hot-air post-treatment, the treated samples were wrapped and incubated at 100 °C in a convection oven. The samples for leaching tests were randomly taken after 0.5, 1, 3, and 6 h.

Retention Analysis

The retention of Cu (expressed as CuO) and DDAC was measured for wood treated with control ACQ and reformulated preservatives. For CuO retention, wet ashing procedures were applied as described in the AWPA A7-04 (2015) standard. The DDAC retention in the treated wood specimens was measured according to the Korean Forestry Promotion Institute's standard test method for wood products.

Sawdust from the treated wood was first prepared using a Wiley mill and screened to 20-mesh. A total of 1 g of sawdust for CuO retention was digested using the peroxide-nitric acid method, and the metal concentration in the resulting solution was analyzed using inductively coupled plasma-optical emission spectrometry (ICP-OES), as described in the AWPA A20-03 (2016). For DDAC retention, 1 g of sawdust was placed in 50 mL of absolute ethanol and subjected to ultrasonic extraction. The concentration of DDAC in the extract was then quantified using high-performance liquid chromatography with ultraviolet detection (HPLC-UV). The HPLC column used was a C_{18} column (inner diameter 4.6 mm, length 250 mm, particle diameter 5 µm), and the column temperature was set at 40 °C. A mixture of methanol and buffer at a ratio of 23:77 was used as the mobile phase. The buffer solution was prepared by combining 1.56 g of sodium dihydrogen phosphate, 20 mL of 25 mM sodium 2-naphthalenesulfonate solution, and the calculated volume of 10% phosphoric acid in a 1-L volumetric flask. Distilled water was then added to reach the final volume. The flow rate of the mobile phase was set at 1.0 mL/min.

Laboratory Fixation and Leaching Tests

Fixation tests were performed following the previous methods (Lee and Cooper 2012; Ung and Cooper 2005). The wet-conditioned samples at different times were randomly taken out and squeezed in a press and the squeezed-out liquid was collected. The copper concentrations in this liquid and the treating solutions were analyzed using ICP-OES. The copper fixation (%) of treated wood was determined by comparing the copper concentration in the treating solutions and the squeezed out liquid. The time to stabilization was determined by when the copper fixation (%) plateaued.

Leaching tests were carried out according to the AWPA E11 (2015). For each leaching batch, six blocks were soaked with 300 mL of distilled water, and the leaching solution was replaced after 6, 24, and 48 h and every 48 h thereafter for two weeks. The leachates collected from the total 9 samples were combined for analysis of metal content using ICP-OES. Cumulative losses of active ingredients were evaluated using the samples after time to stabilize the treated samples.

Laboratory Tests with Biological Efficacy Towards Brown Rots and Termite

Brown-rot resistance of wood treated with commercial ACQ, and reformulated preservatives was evaluated according to the Japanese Industrial Standard (JIS) K 1571 (2004) test method for wood preservatives. In a glass bottle, 350 g of sea-sand was mixed with 100 mL of a medium composed of 4.0% glucose, 0.3% peptone, and 1.5% malt extract by weight. The sea-sand culture with medium was autoclaved at 121°C for 20 min. The inoculum of two brown-rot fungi, *Gloeophyllum trabeum* and *Fomitopsis palustris*, was prepared by growing them in 100 mL of 2% malt extract media. After mechanical homogenization of mycelia in media, 5 mL of inoculum was aseptically spread over the surface of the sterilized sea-sand culture and incubated at 28 °C until the mycelia completely covered the surface. A plastic mesh was then placed on the surface of the sea-sand culture to ensure direct contact with the mycelia. Sterilized wood specimens, including untreated wood and wood treated with commercial and modified ACQ, were placed on top of the mesh and subjected to a decay test at 28 °C for 12 weeks. The percentage mass loss was obtained through comparing the dried weight of each specimen before (m_0) and after (m_1) tests.

$$\text{Mass loss (\%)} = \frac{m_0 - m_1}{m_0} \times 100 \tag{1}$$

The termite resistance of commercial ACQ and reformulated preservatives was evaluated according to the JIS K 1571 (2004) test method for wood preservatives. Acrylic cylinders (8 cm in diameter, 6 cm in height) were used as termite incubation containers. The bottom of each cylinder was sealed with a 5-mm-thick layer of dental plaster. A 2-cm-thick layer of water-soaked cotton was then placed at the bottom. A single wood specimen with a dimension of 20 mm × 20 mm × 10 mm sterilized with ethylene oxide gas was placed in the center of each container. A total of 200 worker termites and 2 soldier termites (*Reticulitermes speratus kyushuensis*) were introduced into each container. The containers were covered with aluminum foil to prevent moisture evaporation and were kept in a controlled environment at 28 °C and over 70% relative humidity for 4 weeks. After 4 weeks, the termite-attacked specimens were cleaned of any surface debris and dried at 50 °C until reaching a constant weight. Through comparing the dried weight of each specimen before and after termite attack, the percentage mass loss was obtained, similar to the method used in the brown rot tests. Further, the number of dead termites in each container was counted to evaluate the mortality rate (%).

Simulated Outdoor Exposure Test

The simulated outdoor exposure test was conducted for evaluating the performance of commercial ACQ and reformulated preservatives against brown-rot and soft-rot fungi. The soil bed method was conducted as described in the AWPA E14-15 (2015). Samples with a dimension of 5 mm × 19 mm × 150 mm were prepared and treated with the abovementioned preservatives. Plastic boxes measuring 26 × 36 × 24 cm^3 were prepared with four drainage holes at the bottom. A 2.5-cm-deep drainage layer of gravel was placed at the bottom, followed by a 19-cm-deep soil layer. The soil used for the test was collected from the Chuncheon experimental site of the National Institute of Forest Science. The soil moisture content for the brown-rot and soft-rot test beds was adjusted to 60% and 100% of the soil's water holding capacity, respectively. A total of 12 wood specimens were inserted into each container, with 75% of their length buried in the soil bed. The boxes were then placed in a controlled environment at 28 °C and 85% relative humidity. The soil moisture content in the box was monitored weekly, and water was added if needed. The preservative performance in the simulated outdoor exposure test was evaluated by calculating the percentage mass loss, similar to the method used in the brown rot tests.

RESULTS AND DISCUSSION

ACQ Reformulation Effects on Copper Leaching

Table 1 shows the average retention (CuO + DDAC) of wood treated with control ACQ and reformulated preservatives. The CuO retention in the control ACQ-treated wood was 2.7 kg/m^3. The CuO retention of treated wood gradually decreased as the copper proportion decreased in the formulation.

Table 1. Retention of Wood Treated with Control and Reformulated Preservatives

Formulation	CuO (kg/m^3)	DDAC (kg/m^3)	ACQ (CuO + DDAC) (kg/m^3)
Control ACQ 2:1 = CuO:DDAC Cu:Mea = 1:4	2.7 (0.2)	1.4 (0.1)	4.1
10% copper reduction Cu:Mea = 1:4	2.5 (0.3)	1.7 (0.1)	4.2
20% copper reduction Cu:Mea = 1:4	2.2 (0.3)	2.0 (0.1)	4.2
30% copper reduction Cu:Mea = 1:4	1.9 (0.1)	2.2 (0.1)	4.1
40% copper reduction Cu:Mea = 1:4	1.7 (0.1)	2.6 (0.1)	4.3
40% copper reduction Cu:Mea = 1:2.75	1.8 (0.1)	2.5 (0.1)	4.3
All values here represent the average of 5 replicates and ones in parentheses are the standard deviations			

The increased DDAC proportion in the formulation compensated for the CuO reduction, thereby leading to the average retention of wood treated with reformulated preservatives remaining in a range of 4.1 to 4.3 kg/m³ and satisfying the retention requirement for aboveground use (2.4 kg/m^3) according to AWPA U1-24 (2024).

Table 2 shows time to copper stabilization, maximum copper fixation (%), and cumulative copper and DDAC leaching (%) of wood treated with control ACQ and reformulated preservatives. The reduction of copper content in a preservative formulation improved the copper fixation properties of treated wood (Table 2). As the copper content decreased, the time to copper stabilization generally shortened, and the maximum copper fixation increased. The time to copper stabilization shortened from 15 to 6 days when the copper content in the formulation decreased 30% or more. The maximum copper fixation barely showed a difference between wood treated with control ACQ and a preservative of formulation with a 10% reduction in copper content. However, notable differences in fixation were observed in formulations with a reduction of 20% or more in copper content. The maximum copper fixation of wood treated with the preservative with a 40% reduction in copper content was 93.7%, which is 5.4% higher than the 88.3% of wood treated with control ACQ. Consistent with fixation properties, cumulative copper leaching also decreased from 6.9% to 4.9%, which corresponded to 4130 mg/m^2 and 1911 mg/m^2, respectively.

The preservative with a 40% reduction in copper content was further reformulated by adjusting a molar ratio of Cu to Mea from 1:4 to 1:2.75. Lowering the solvent proportion enhanced the copper leaching resistance of treated wood (Table 1).

Table 2. Effects of Lower Copper Content and Mono-Ethanolamine in a Preservative Formulation on Copper Fixation and Leaching

Formulation	Time to Copper Stabilization (days)	Maximum Copper Fixation (%)[a]	Cumulative Copper Leaching (%)[a]	Cumulative DDAC Leaching (%)[a]
Control ACQ	15	88.3 (1.1) A	6.9 (1.1) A	0.1 (0.0) A
10% copper reduction Cu:Mea = 1:4	10	91.2 (2.6) AB	5.9 (0.6) A	0.1 (0.0) A
20% copper reduction Cu:Mea = 1:4	10	92.5 (1.5) B	5.4 (0.4) A	0.1 (0.0) A
30% copper reduction Cu:Mea = 1:4	10	92.7 (1.2) B	5.4 (0.3) A	0.1 (0.0) A
40% copper reduction Cu:Mea = 1:4	6	93.7 (1.5) B	4.9 (0.4) B	0.1 (0.0) A
40% copper reduction Cu:Mea = 1:2.75	6	94.1 (0.1) B	3.0 (0.2) B	0.1 (0.1) A

[a] Values represent the average of triplicates and ones in parentheses are the standard deviations.
*Numbers followed by the same letter in each column are not significantly different (α = 0.05) according to Duncan's multiple range test.

The preservative with a molar ratio of Cu to Mea of 1:2.75 exhibited no difference in time to copper stabilization and maximum copper fixation compared to the preservative with a molar ratio of Cu to Mea of 1:4. However, in laboratory leaching tests, the cumulative copper leaching decreased from 4.9% to 3.0%, which corresponded to 1911 mg/m^2 and 1170 mg/m^2 in amounts, respectively. The cumulative leaching of DDAC was less than 0.1% for all formulations and exhibited no significant difference between the groups, indicating that lowering the proportion of both copper and Mea had no effect on DDAC leaching.

Improved fixation properties in wood treated with a preservative of lower copper content might be explained by two reasons. Firstly, as the cation exchange capacity of wood for Cu-Mea adsorption is limited. Thus, a reduced copper content in a formulation can increase the percentage of copper fixation. Secondly, keeping the same molar ratio of Cu:Mea, the absolute content of Mea in a preservative decreases with a reduction in copper content. Less buffering capacity by Mea in a solution can cause a rapid and greater pH drop within treated wood. A faster and greater pH decrease is favorable for physical precipitation of copper, thereby shortening the time to stabilization and increasing the maximum copper fixation. Figure 1 shows the pH change of preservatives squeezed out of treated wood at different times. These results directly indicate that the pH drop was faster and greater in wood treated with the preservative formulated with reduced copper content. Further, a preservative formulated with a molar ratio of Cu to Mea of 1:2.75 within wood showed a lower pH value compared to other formulations, indicating that the Mea adjustment also induced favorable conditions of physical precipitation and resistance to leaching of copper from treated wood.

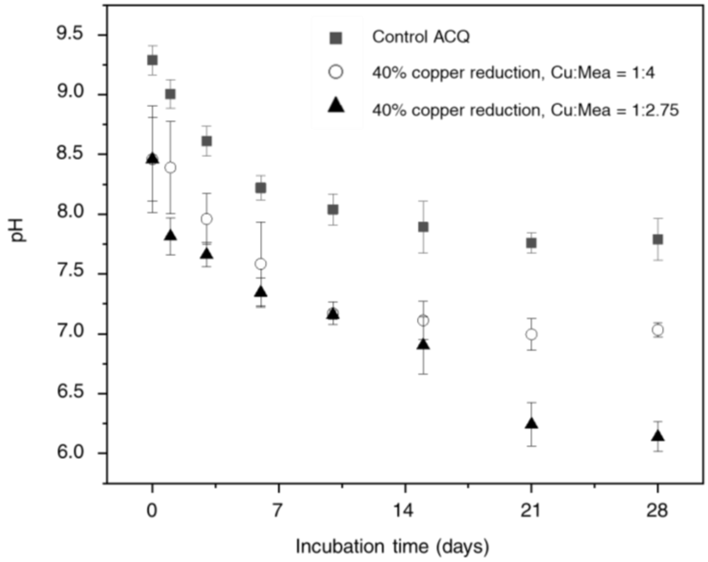

Fig. 1. pH changes of control ACQ and reformulated preservatives within treated wood as a function of incubation time

Hot air post-treatment was applied to wood treated with a preservative formulated with a 40% copper reduction and a molar ratio of Cu to Mea of 1:2.75. Table 3 shows the cumulative copper leaching (%) from the treated wood after post-treatment with different durations. With just a simple 30 min of post-treatment, cumulative leaching was only 2.0%, which was almost half of the samples wet-conditioned at 21 °C for 6 days (Table 1). Increasing time of post-treatment time could not further improve leaching resistance. These results indicated that hot air post-treatment reduced the time to copper stabilization of treated wood and enhanced copper leaching resistance. This faster copper stabilization is likely due to the copper conversion from soluble cupric ions to insoluble cuprous precipitates (Yu *et al.* 2009).

Table 3. Effects of Hot Air Treatment on Copper Leaching from Wood Treated with a Preservative with a 40% Copper Reduction and a Molar Ratio of Cu to Mea of 1:2.75

Post-treatment Time (h)	Cumulative Cu Leaching (%)[a]
0.5	2.0 (0.1) A
1	2.0 (0.1) A
2	2.1 (0.1) A
3	2.1 (0.1) A
6	2.2 (0.3) A

[b] Values represent the average of triplicates and ones in parentheses are the standard deviations.

*Numbers followed by the same letter in each column are not significantly different (α=0.05) according to Duncan's multiple range test.

Biological Efficacy of Reformulated Preservatives

Biological efficacy of two reformulated preservatives was evaluated compared with commercial ACQ; 1) a preservative formulated with a 40% copper reduction and a Cu:Mea ratio of 1:4, 2) a preservative formulated with a 40% copper reduction and a Cu:Mea molar ratio of 1:2.75. Laboratory decay tests, termite resistance, and simulated outdoor exposure were conducted.

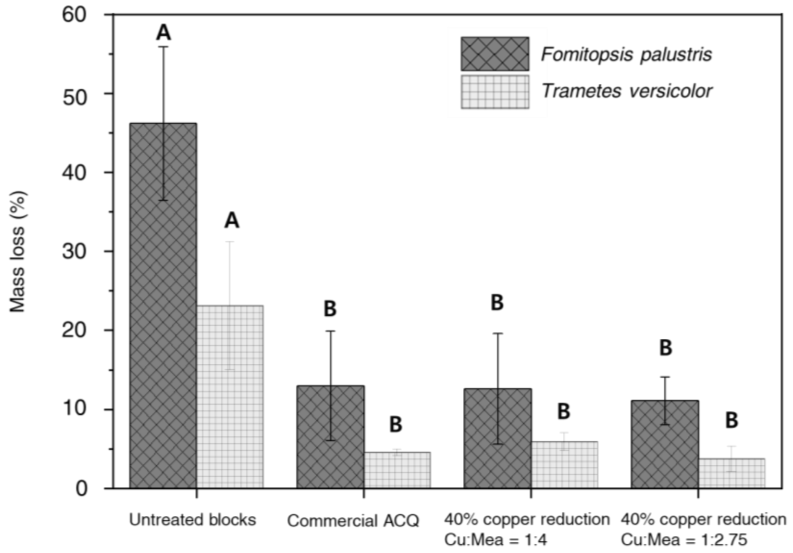

Fig. 2. Brown-rot and white-rot resistance of wood treated with commercial ACQ and reformulated preservatives. Bars represent the average values from ten replicates. Significant differences among preservative groups are indicated by different letters over error bars ($p < 0.05$).

Figure 2 shows the mass loss of treated wood against one brown-rot fungus, *F. palustris*, and the white-rot fungus, *Trametes versicolor*, *F. palustris*, and *T. versicolor.* The test results showed average mass losses of 46.2% and 23.1% of untreated blocks, respectively, which satisfied the requirement for decay test validity of JIS K 1571 (2004). For both fungi strains, the mass loss of the wood treated with the reformulated preservatives showed no significant differences compared with commercial ACQ, suggesting that the reformulated preservatives have comparable biological efficacy with commercial ACQ. It should be noted that the higher mass losses caused by *F. palustris* compared to *T. versicolor* in treated blocks can be attributed to its copper resistance.

Figure 3 presents the mass loss of treated wood against termite attack. The mass loss (%) of the untreated blocks was 23.1%, in which the termites exhibited only 32% of mortality. These results confirm the validity of termite resistance test described in JIS K 1571 standard (2004). No statistically significant difference in mass loss was observed among commercial ACQ and reformulated preservatives, and the termites introduced to commercial ACQ and reformulated preservatives were fully dead. These results suggest that the biological efficacy of reformulated preservatives against termites is equivalent to that of commercially available ACQ.

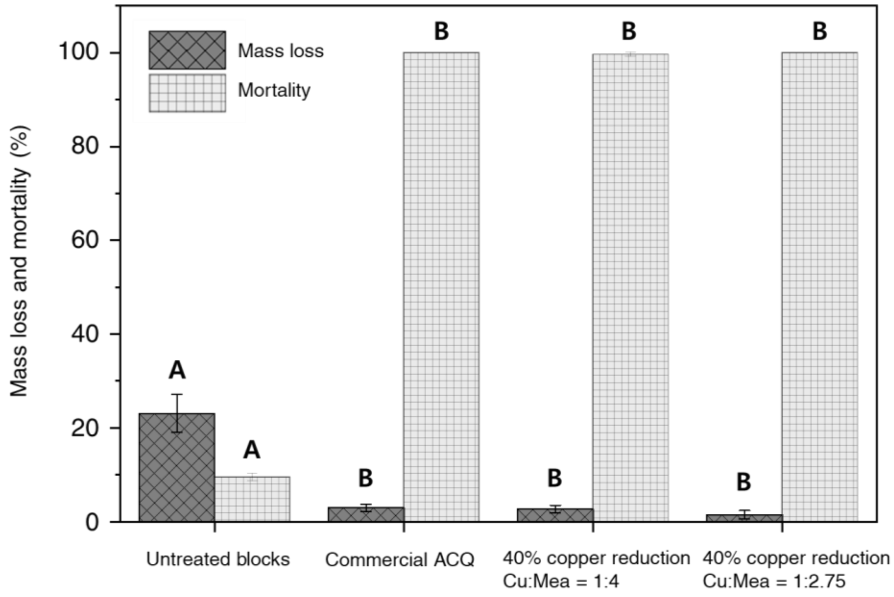

Fig. 3. Termite resistance of wood treated with commercial ACQ and reformulated preservatives. (A) Mass loss (%) and (B) Mortality (%) of termites after termite tests. Significant differences among preservative groups are indicated by different letters over error bars ($p < 0.05$).

To deeply evaluate the biological efficacy of reformulated preservatives, simulated outdoor exposure tests were conducted following the AWPA E14-15 (2015). The retention level of samples used were prepared to satisfy the required retention level specified in AWPA E14-15. In addition, fungal strains were isolated following the method of a previous study (Heo *et al.* 2019), revealing four genera of basidiomycetes and seven genera of ascomycetes that may cause soft rot (Appendix). This information confirmed that the soil harvested was suitable for the simulated test. Figure 4 indicates the mass loss of wood treated with commercial ACQ and reformulated preservatives. After three months of exposure, no mass loss was observed in the specimens treated with three types of preservatives. This indicates that the mock outdoor exposure test is proceeding normally. However, the untreated specimens showed mass losses of 2.0% and 1.3% by basidiomycetes and ascomycetes attack, respectively. White mycelia were observed on some untreated specimens used for the basidiomycete evaluation, and severe discoloration was found on some untreated specimens used for the ascomycete evaluation (Appendix).

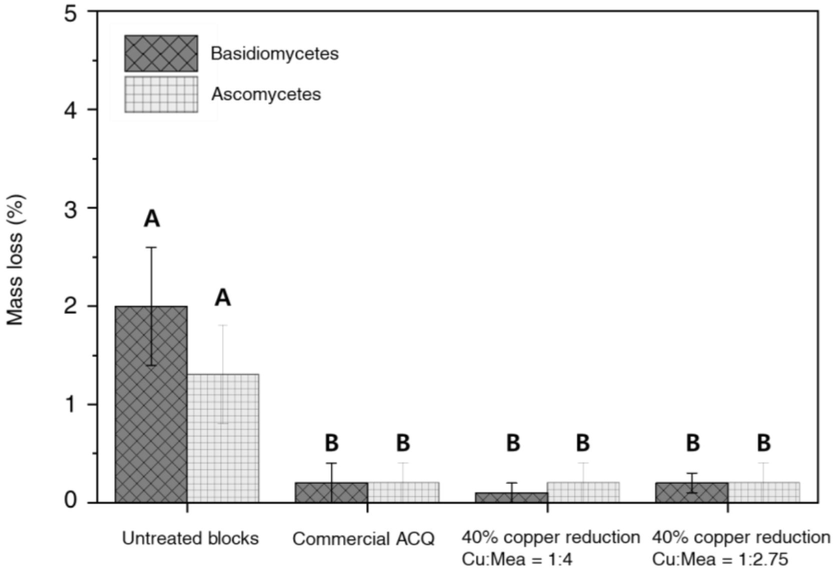

Fig. 4. Basidiomycetes and ascomycetes resistance of wood treated with commercial ACQ and reformulated preservatives, subjected to simulated outdoor exposure test. Significant differences among preservative groups are indicated by different letters over error bars ($p < 0.05$).

Comparable biological efficacy of two reformulated preservatives might be due to higher DDAC proportion. The DDAC compensated for reduced CuO within treated wood might provide equivalent efficacy against fungi and termites. Butcher *et al*. (1977) reported that wood solely treated with DDAC, at a retention of 1.6 to 3.2 kg/m³, exhibited noticeable preservative performance against the brown-rot fungi *Gloeophyllum trabeum* and *Poria placenta*, as well as the white-rot fungus *Fomes gilvus* (Butcher *et al.* 1977). Robinson and Laks (2010) showed that *Postis placenta* failed to colonize DDAC treated wood with a retention level of 0.1 kg/m³ or more. Hwang *et al*. (2006) reported that wood treated with 0.5% DDAC (retention: 3.0 to 3.99 kg/m³) as a stand-alone preservative exhibited high termite resistance against *Coptotermes formosanus*. Considering lower toxicity to mammalian cells and strong leaching resistance to aquatic settings, a formulation with lower copper and higher DDAC proportion might be promising.

Further study is required to evaluate the long-term performance of reformulated preservatives. Even with enhanced copper leaching resistance, the preservative performance may change with extended exposure due to potential leaching, migration, and degradation of DDAC. It is well known that quats, including DDAC, can easily migrate to soils, and they are highly susceptible to biodegradation by various soil microbes. A previous study showed that 8.3 to 34.7% of synthesized quats in treated wood naturally migrated to soil. Most types of quats exhibited susceptibility to biodegradation by fungal species, leading to the lower biological efficacy of treated wood (Zabielska-Matejuk and Czaczyk 2006).

CONCLUSIONS

1. A preservative reformulation with a reduction of 40% copper content and adjustment of molar ratio of Cu:Mea from 1:4 to 1:2.75 improved copper fixation and leaching properties of alkaline copper quat (ACQ) preservative for wood.
2. A 30-min, hot air post-treatment at 100 °C could improve copper leaching resistance.
3. The reformulated preservative has comparable biological efficacy in comparison with commercial ACQ because the higher didecyldimethylammonium chloride (DDAC) proportion compensates for the CuO reduction.

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APPENDIX

Table S1. Fungal Species Isolated from the Soil Used for Simulated Outdoor Exposure Test

Fungal Type	Fungal Species
Basidiomycetes	*Hapalopilus* sp.
Basidiomycetes	*Fuscoporia* sp.
Basidiomycetes	*Peniophora* sp.
Basidiomycetes	*Entomocorticium* sp.
Ascomycetes	*Chloridium aseptatum*
Ascomycetes	*Exophiala tremulae*
Ascomycetes	*Hazslinszkyomyces aloes*
Ascomycetes	*Metarhizium robertsii*
Ascomycetes	*Ochroconis* sp.
Ascomycetes	*Penicillium* sp.
Ascomycetes	*Saitozyma podzolica*

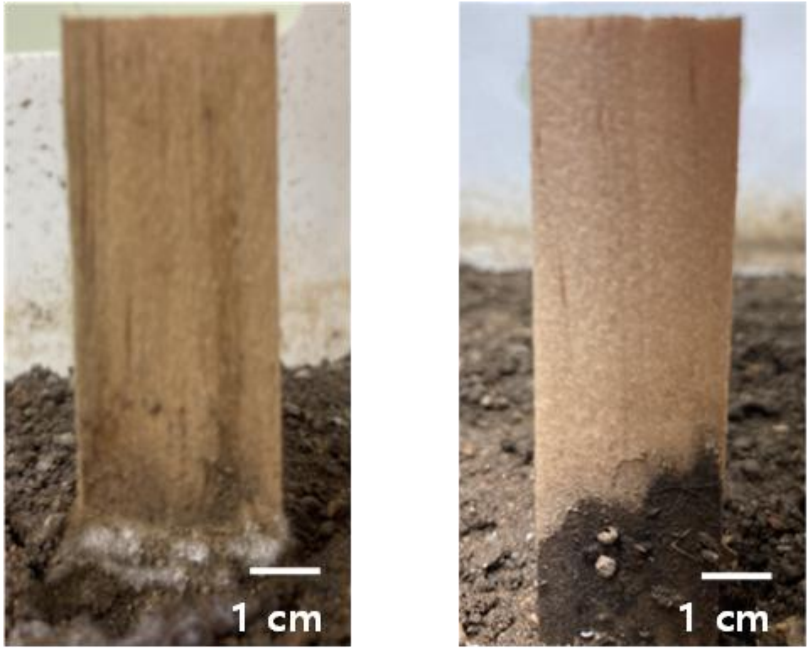

Fig. S1. Untreated wood samples buried in a soil-bed for basidicomycetes (left) and ascomycetes (right), respectively.